DASA 539 B

HASP

Second Special Report on

High Altitude Sampling Program

Defense Atomic Support Agency

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1 AUGUST 1961

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TECHNICAL ANALYSIS REPORT

DASA-539 B

SECOND SPECIAL REPORT ON THE HIGH ALTITUDE SAMPLING PROGRAM (HASP)

by

Major Albert K. Stebbins, III

RADIATION DIVISION

This document was previously published as DASA 539S "Second Special Report to the Government of Argentina".

1 August 1961

HEADQUARTERS, DEFENSE ATOMIC SUPPORT AGENCY
WASHINGTON 25, D. C.

ABSTRACT

This document is the second special report of the High Altitude Sampling Program (HASP). DASA-532 was the first special report. A recalibration of the U-2 ducts has diminished the previous discrepancy noted in the flow rates. Absolute values of radionuclide concentrations are increased 8 to 13% by this recalibration. A discussion on the structure and nature of the 0.1 to 1.0 micron family of naturally occuring stratospheric aerosol is presented. This dust layer of ammonium sulfate appears to be generated in the stratosphere and may play an important role in lower stratospheric fallout processes. A detailed discussion of stratospheric concentrations of a number of nuclides is presented. Sr-90 and W-185 inventories and distributions from August 1957 to May 1960 are discussed. By May 1960 maximum tungsten values are found in the lower altitude equatorial regions while maximum strontium values are found in the higher altitude polar regions. The tungsten stems from low altitude HARDTACK shots only while the strontium appears to show influx from Teak and Orange. The tungsten and strontium concentration variations shed considerable light on stratospheric mixing processes. Ba-140, Sr-89, and Ce-144 concentrations assist in determining the age of debris and thus allow following of stratospheric mixing and transfer processes. A number of definite seasonal effects are noted. Transfer from the tropics to the polar regions is greatest during the winter and all but ceases during the fall. Various possible modes of tropospheric-stratospheric interchange are discussed. A detailed discussion of fallout from Teak and Orange debris is presented. Rh-102 data suggests at least 10% of Orange was in the lower stratosphere by May 1960. Ce-144 and Sr-90 data suggests that 25% of the debris in the polar regions in early 1960 was from Teak and Orange. A half residence time of about 5 years in the mesosphere for Teak and Orange is suggested. Entry into the lower stratosphere apparently proceeds through rapid downward mixing in the polar regions during the winter night. A study of natural radionuclides in the stratosphere is presented and assists in measurement of stratospheric processes. Elements studied include C-14, H-3, Pb-210, Be-7 and P-32. Be-7 and P-32 concentrations in the stratosphere are about that expected from cosmic ray production. Pb-210 in the stratosphere may partially result from equatorial bomb tests. Comments are made on surface fallout measurements which corroborate the HASP measurements. Seasonal and latitudinal effects are noted. The contribution of French tests are calculated. The hazard of radioactive fallout is assessed by nuclide and dose type. The 30 year genetic or whole body dose in the U. S. from Cs-137 and elements of shorter half life is shown to be less than 100 millirem or less than 3% of the natural background. The lifetime, 70 year, somatic bone dose to children in the U. S. is shown to be about 200 millirem of which half is from Sr-90. This is less than 2% of the population MPD. Finally several appendices provide U-2 operational scenes, useful constants and conversion factors, and a summary of nuclear detonations.

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ACKNOWLEDGEMENTS

The HASP program has been an extremely diverse and extensive project and has been the culmination of the efforts of a broad spectrum of workers including scientific, operational, diplomatic, and administrative as well as other planning and support personnel. While it is thus impossible to acknowledge adequately the assistance rendered by everyone who contributed to the success of the program, a few may be singled out. The recalibration of the U-2 ducts was conducted and supported by Dr. Hugh Dryden, Dr. Smith de France, and Mr. George Holden of NASA; Prof. Elliott G. Reid of Stanford University; and Capt. Budd Knapp of the Air Force Flight Test Center. The major analytical program was conducted by Dr. Johannes A. Van den Akker of the Institute of Paper Chemistry; and Dr. Herbert W. Feely, Dr. Jerome Spar, Dr. Philip W. Krey, Dr. Alan Walton, Dr. James P. Friend and Dr. J. Laurence Kulp of Isotopes, Inc. Helpful assistance and interpretive comment was offered by Dr. Lester Machta, Mr. Robert J. List and Mr. Kosta Telegadas of the U. S. Weather Bureau; Mr. Joshua Z. Holland and Dr. Harold A. Knapp of the U. S. Atomic Energy Commission; Dr. Edward A. Martell, Dr. Christian E. Junge, Dr. Marvin I. Kalkstein and Mr. James E. Manson of the Air Force Cambridge Research Laboratory; Dr. Luther B. Lockhart, Jr., of the U. S. Naval Research Laboratory; and Dr. Willard F. Libby of the University of California. Finally, special thanks are due to the crews and support personnel of the Military Air Transport Service, the Strategic Air Command, and the Air Research and Development Command who obtained the necessary samples under conditions which could be overcome only by the acme of performance.

NOTE

This document was previously issued under the title, "Second Special Report to the Government of Argentina". Several minor corrections have been made at various places in the text. In addition, a short section has been appended to Chapter X which discusses HASP soil studies and another to Chapter XI which discusses possible biological exposure from future tests. It should be pointed out that all conclusions in this report are based on weapons testing conducted prior to 1 August 1961. Several calculations were made on the assumption that no further atmospheric testing would be conducted after that date.

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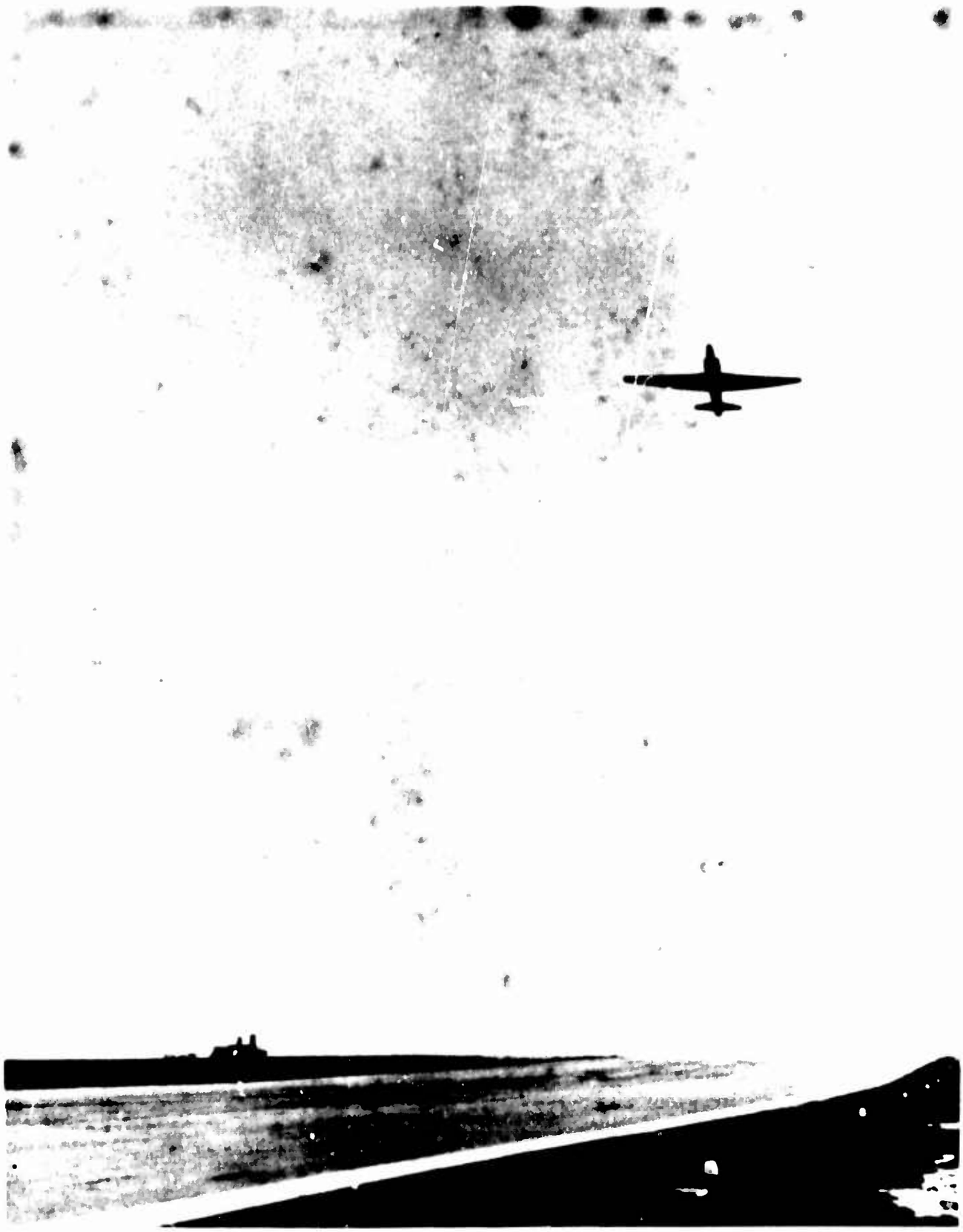


Chapter I

INTRODUCTION

Background

Since 1957, the Defense Atomic Support Agency has conducted an atmospheric sampling program using U-2 aircraft. Known as the High Altitude Sampling Program (HASP), its purpose has been to determine the role played by the stratosphere in the world-wide distribution of fission products from nuclear explosions. HASP has been an integral part of the coordinated program of study of atmospheric radioactivity which is being conducted by the United States Government[1].*

The purpose of this publication is to report some of the observations made during Phases IV and V of HASP and the conclusions to be drawn from them. Since June 1960, the semiweekly collection schedule has been curtailed and the DASA program has merged with studies being jointly conducted by the U. S. Atomic Energy Commission, the U. S. Weather Bureau, and the Air Force Cambridge Research Laboratory (AFCRL). Semiannual spot checks at various places in the world are being conducted under this integrated program. Analyses of the samples obtained are conducted at several laboratories, both contract and government. Radiochemical analyses as well as particle size distribution and particle composition studies are in progress. Anton[1] has reported the broad scope of the overall program.

The details of operation of the HASP meridional network and the analytical procedures used to obtain the HASP data have been outlined in the previous special report (DASA 532) and will not be repeated here. Some of the results previously reported[2-10] will be included to provide some continuity to the reader. A brief resumé of the results obtained since 1 January 1960 and the contents of this

* All references are listed in Appendix IV.

report follows.

Recalibration of Ducts

During the course of analyses of filter papers taken from the nose and hatch ducts on the U-2 aircraft, a noticeable discrepancy appeared in the activity of simultaneously exposed samples. It was felt that this discrepancy (about 20% more activity on the hatch papers) was due to an improper calibration of the air flow rates through the ducts. A recalibration program was completed which produced new flow rates in a direction which has tended to cut the discrepancy in half. In addition, the inventories previously described are about 13% too low. At the present time all the flow rates for papers collected since 1957 are being recalculated. The final report on the HASP program (DASA 1300 in about six volumes, to be published in the fall of 1961) will contain all of the HASP data with recalibrated flow rates. An attempt will be made in this report (DASA 539) to distinguish clearly between data that has resulted from recalculated duct flows and that which has not. In general, all data from Phase V result from recalculated flow rates while data from previous phases do not.

Stratospheric Aerosol

Dust particle collections suitable for study with electron microscopes and with electron microprobes have been made since January 1960. These studies made by Friend[11] at Isotopes, Incorporated, and by Junge, et. al.[12], at AFCRL, have confirmed previous balloon investigations[13] which show a layer of stratospheric aerosol with a maximum concentration at about 60,000 feet. The composition appears to be a sulfate of ammonia produced in the stratosphere by the action of ozone or sunlight on hydrogen sulfide and sulfur dioxide. These particles may play an

important role in scavenging of radioactive particles from the lower stratosphere. This report will contain a discussion of these results and their implication on stratospheric residence times.

Radiochemical Analyses

Since one of the major goals of HASP has been to obtain the stratospheric distribution of Sr-90 as a function of time, these data will be reported along with a discussion of their significance. Tungsten-185 injected into the tropical stratosphere in 1958 provided a unique tracer to follow a test series without obscuration by other tests. These data and their implications will be reported. A residence half-time of less than one year for lower tropical stratospheric injections is suggested by these data with movement out of the tropical stratosphere by turbulent diffusion[9] rather than "Brewer-Dobson"[14,15] circulation. Incursions of Sr-90 from Teak and Orange, the two high altitude megaton bursts of the HARDTACK test series, are noted in both the Northern and Southern hemispheres with the maxima occurring at the highest altitudes and highest latitudes sampled. Another tracer, Rh-102, produced in the Orange shot is detected and allows a measure of the residence time of debris in the mesosphere to be calculated.

Other fission product nuclides such as Sr-89, Ce-144, and Cs-137, as well as plutonium, have been detected. Use of some of these nuclides to determine the age of debris in the stratosphere assists in describing the mixing and transfer of the debris from place to place within the stratosphere and into the troposphere. A fairly well documented picture as emerged which allows a selection from among the various theories of stratospheric mixing which have been proposed.[9,15,16]

Certain radionuclides such as Be-7, P-32, C-14, H-3, and Pb-210, occur as natural by-products of cosmic radiation and decay of radon. Some are also produced

in nuclear detonations. Concentrations of these nuclides have been measured both in the stratosphere and at sea level. A discussion of their distribution sheds additional light on the stratospheric and tropospheric mixing processes.

Surface Measurements

A number of studies of radioactivity in rain and air samples collected at the surface of the earth have been undertaken by many investigators since 1954. The major fraction of the fission products measured in these programs has undoubtedly come from nuclides which were originally injected into the stratosphere. Many of the results of these surface programs are, consequently, directly related to stratospheric processes. An attempt will be made to compare some of these results with HASP results. One important phenomenon, namely, the apparent rise in rate of fallout during the springtime, will receive considerable attention.

Biological Hazard

One goal of fallout measurement under the HASP and other programs has been the assessment of the biological hazard associated with fallout. Sufficient time has elapsed since the last large testing series and sufficient measurements have been made in a variety of samples and locations to make rather accurate estimates of the biological doses to be expected from the past nuclear weapons tests. The most significant isotopes as far as internal irradiation is concerned include Sr-90, Cs-137, I-131, C-14, and possibly plutonium. For external exposure, the significant isotopes are Cs-137, Zr-95, Ru-103, Ru-106 and Ce-144. An evaluation of the amount and type of dose to be expected from each of these isotopes will be presented. While no attempt will be made to minimize the hazard it will be seen that these doses from world-wide fallout are small when compared to the dose received from the ever present natural background of radiation from cosmic rays and rocks.

Chapter II

CALIBRATION OF U-2 DUCTS

Introduction

During the course of the HASP program attempts have been made to determine accurately the flow of air through the filter paper (IPC-1478) used in the sampling ducts of the U-2. Originally a theoretical relationship was used based on an extrapolation of data obtained from the F-33 sampler, a wingtip sampler which can be mounted on T-33 or B-57 aircraft. This sampler had been previously calibrated in a wind tunnel. Since there was some doubt as to the validity of the extrapolation, it was decided to instrument the ducts and measure directly the face pressure and pressure drop across the filter paper under sampling conditions. This program was undertaken at Laughlin Air Force Base, Texas, in 1958. The results of this calibration were reported in DASA-532.

After 1 June 1959, a series of dual samplings using both the nose and hatch samplers on the U-2 was performed. It became increasingly obvious that the activity concentration on the hatch filter papers was about 20% higher than the activity on the nose filter papers. Since the papers were exposed simultaneously, it was expected that equal activities would be obtained. The calibration data for these ducts showed the linear velocity of flow through the nose to be about 20% higher than through the hatch. Since this difference could account for the noted discrepancy, it was decided to recalibrate the ducts (of the same aircraft previously calibrated) under better conditions.

Instrumentation

The nose duct has a circular opening 3.75 inches in diameter, while the hatch duct has an elliptical opening 8.25 by 4.75 inches. (See Fig. 1) When the duct

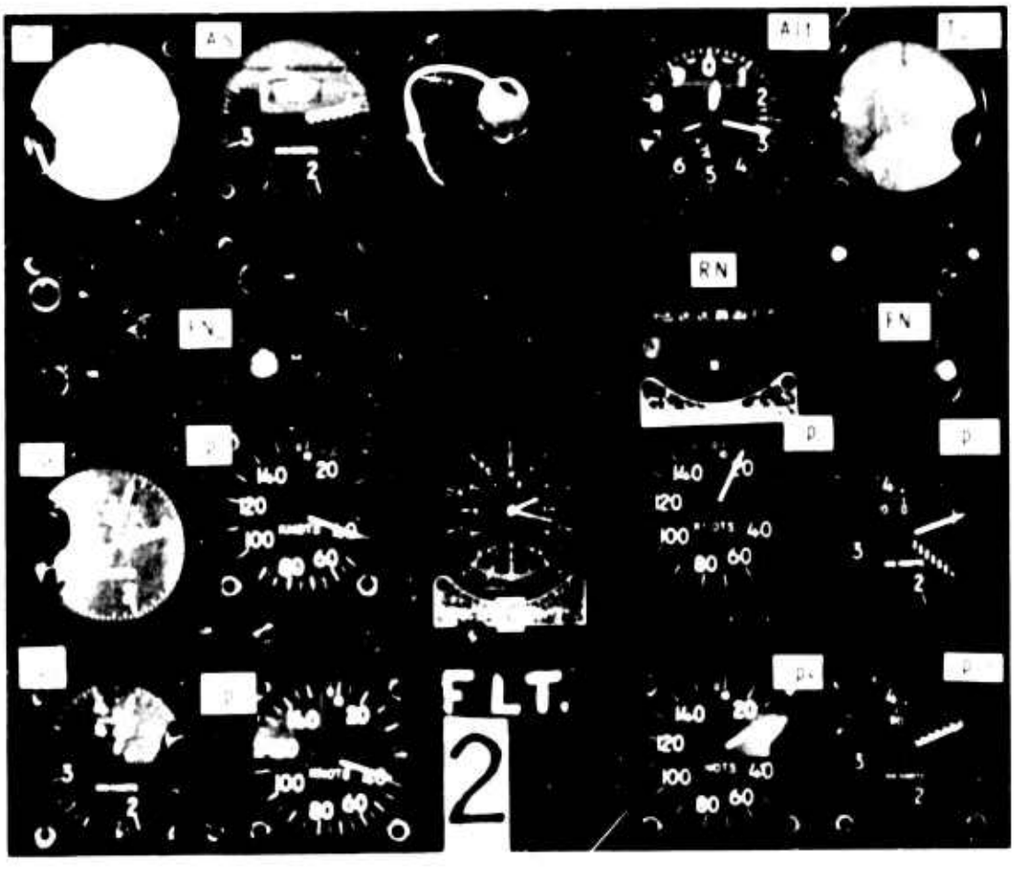
doors are open an obstruction 0.25 inches high remains across the center of the opening. The ducts gradually widen until the filter papers are reached. The effective filter areas are 0.55 and 1.38 ft^2 for the nose and hatch ducts respectively.

An instrument theater supplied by the Ames Laboratory of NASA containing an altimeter, airspeed indicator, clock, 2 temperature guages (free air, theater), and 8 pressure instruments, was installed in the equipment bay of the aircraft above the hatch duct. The instruments were photographed in a mirror by a camera mounted in the plane of the instruments. Pressure lines connected the instruments to the pitot-static system and to small openings in each duct at the entrance and immediately fore and aft of the filter paper. Poppet valves to protect the instruments when not in use were also installed. Figures 2, 3, and 4 show the installation from the face, top, and bottom respectively. Figure 5 shows the arrangement of the plumbing in the recording system. Six of the pressure instruments were actually airspeed meters of the standard drum type and two were helicopter airspeed meters for accurate measurements in the low range.

Results

The calibration flights were carried out under the supervision of Professor Elliott G. Reid, of Stanford University, at Edwards Air Force Base, California, during July 1960. Several flights were made at varying airspeeds and altitudes at light and heavy aircraft weight using precalibrated filter papers of nearly equal bulk density and perforated plates of varying porosity.

Reduction of the data was performed by the method described by Reid[2,17,18]. Basically this amounts to measuring the pressure drop across the filter in flight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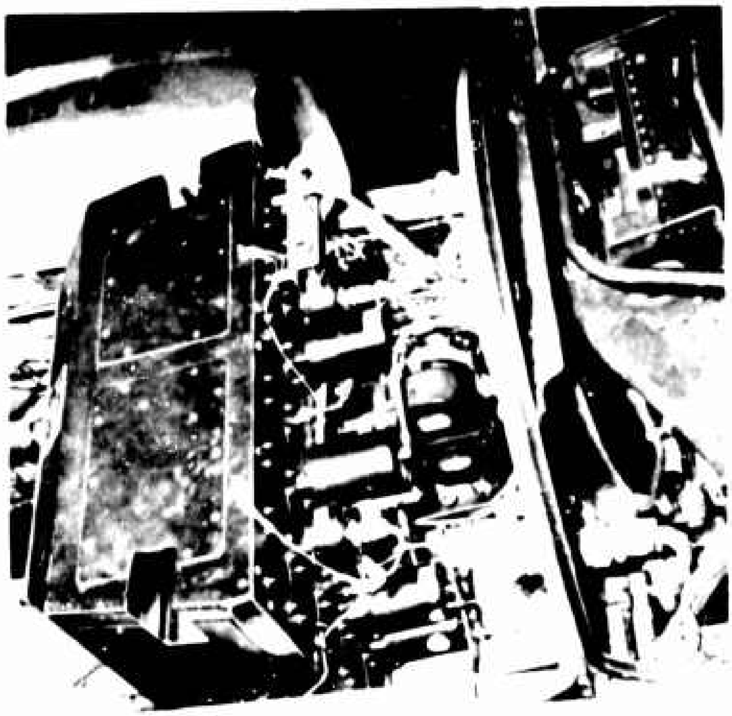
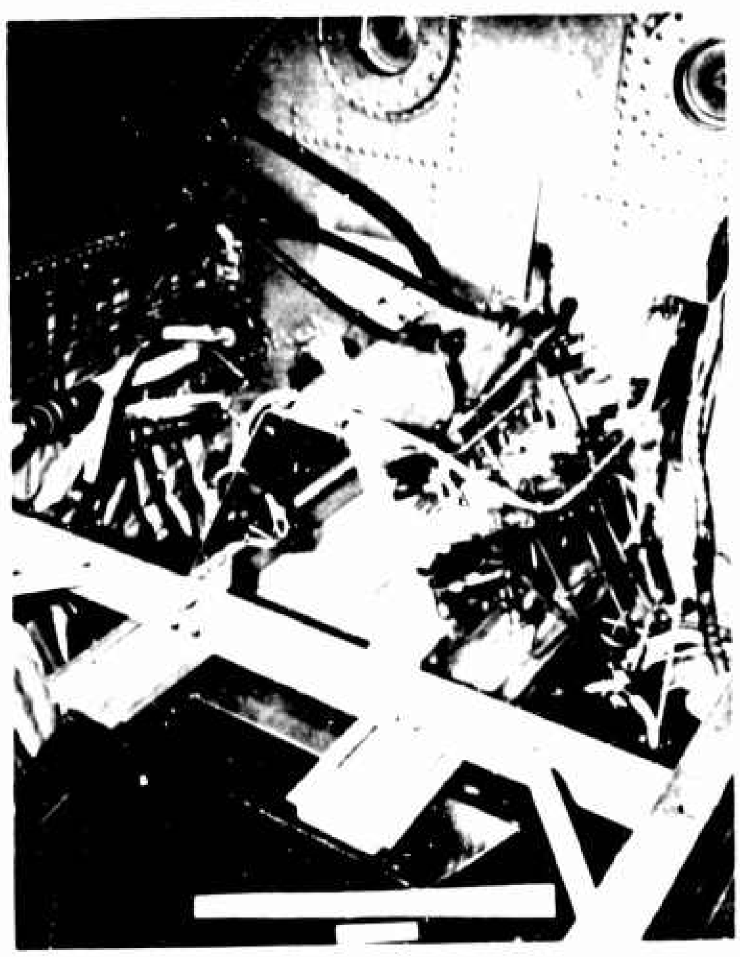
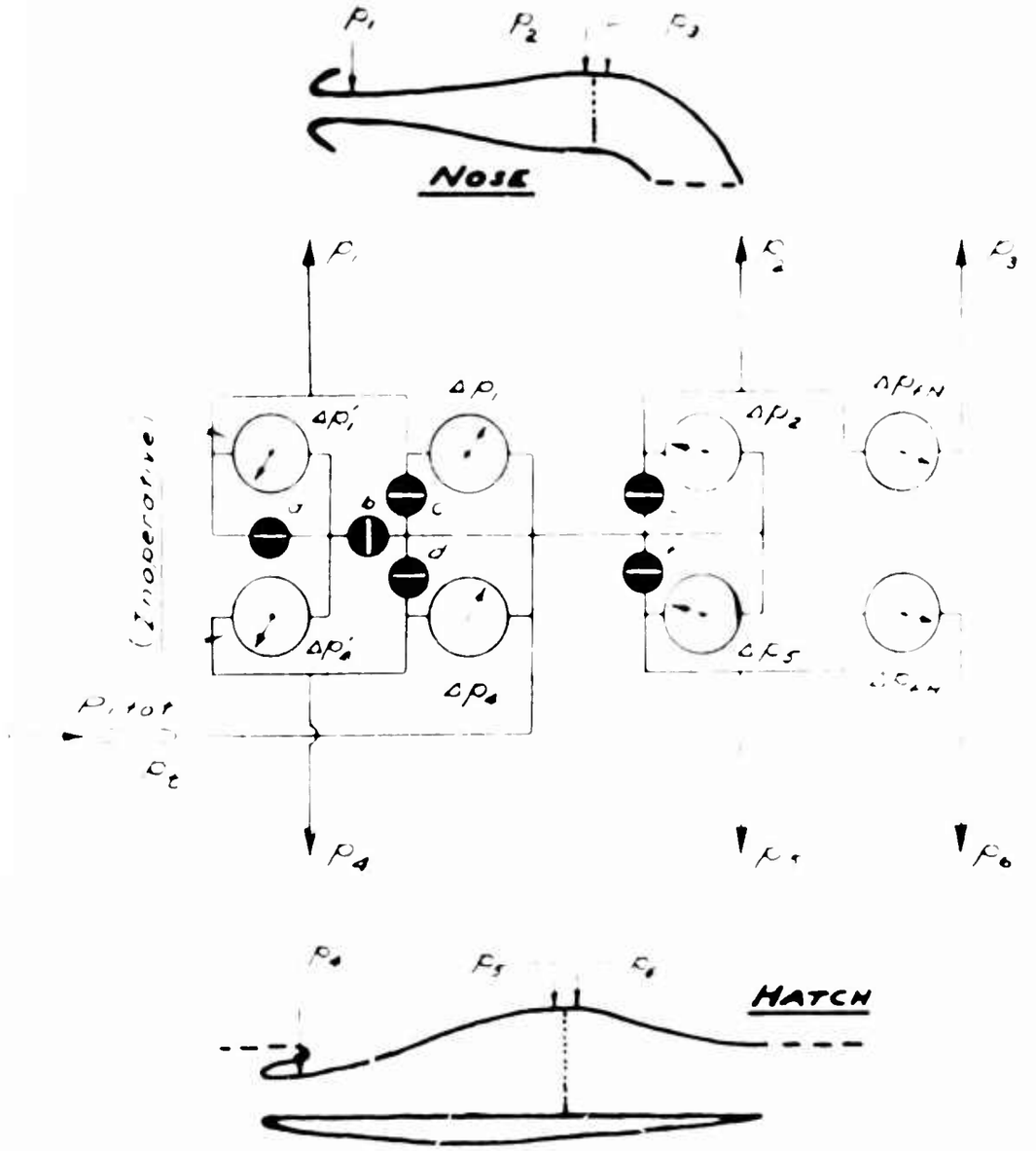




DUCT PRESSURE RECORDING SYSTEM



NORMAL RECORDING CONFIGURATION

p_t = Total pressure

$\Delta p_1 = p_t - p_1$

$\Delta p_2 = p_t - p_2$

$\Delta p_{tN} = p_2 - p_3$

$\Delta p_4 = p_t - p_4$

$\Delta p_5 = p_t - p_5$

$\Delta p_{tN} = p_5 - p_6$

a - Poppet valve normally open - solenoid closed actuated by pilot's switch

b - As in a but normally closed

c, d left - Poppet valves normally open - solenoid closed actuated by entry valve micro-switches

FIGURE 5

and then determining the velocity of flow through the paper from a pressure drop-velocity characteristic curve determined by wind tunnel calibration of the filter paper used in the duct.

The flight data obtained are remarkably consistant attesting to the excellence of the instrumentation. The pressure at the face of the nose filter was about 97% of stagnation pressure and the pressure drop across the filter varied from 70% to 80% of the dynamic pressure. As a result, the velocity of flow through the filter was 70% to 80% of the maximum possible value which would be obtained if the pressure at the face were at the stagnation value and the pressure immediately aft were at the ambient value. This drop in efficiency has two causes; first, the filter paper is sufficiently porous that the flow velocity is about 5% of the free stream velocity so stagnation pressure cannot be reached and, second, there is considerable back pressure in the duct caused apparently by the manner in which it exits at the surface of the aircraft. The nose duct flow rates seemed to be independent of the gross weight of the aircraft, while the hatch duct flow rates showed a change with gross weight of only a few percent.

In the case of the hatch duct, the filter paper face pressures were registered to be consistently lower than those in the nose duct although the aft pressures were the same. As a consequence, the face velocities of the hatch were calculated to be about 10% lower than those of the nose at the usual sampling speed schedule. Radiochemical data [2] indicate that the face velocities should be the same. There are several possible explanations for this difference.

1. The face velocities may really be different and the radiochemical data is in error. Dual filter papers sometimes show as much as 40% difference in activity, although the average difference, using the recalibrated duct data, is 10% more

activity per unit volume sampled by the hatch.* (10% less volume sampled per unit area of filter)

2. The effective areas of the filter papers may not be 0.55 and 1.38 ft^2. This possibility cannot be completely ruled out although autoradiographs show uniform density of activity across the filter papers and selected samples cut from various quadrants at different distances from the center of the paper show no trend in activity density.[19]

3. The pressure at the filter face in the hatch (p_5 in Fig. 5) may have been recorded at a value consistently lower than the actual pressure. It is not known how much the asymmetry of the hatch duct affects the pressure around the periphery of the duct in the vicinity of the filter face. Only one take-off point for the measurement of p_5 was available while a two point manifold situated 180^o apart was available for the same measurement in the nose duct (p_2 in Fig. 5). The nose duct is more symmetrical than the hatch duct. The difference between p_2 and p_5 was about 9% of the dynamic pressure.

4. There may have been excessive leakage around the filter papers past the inflatable seals.

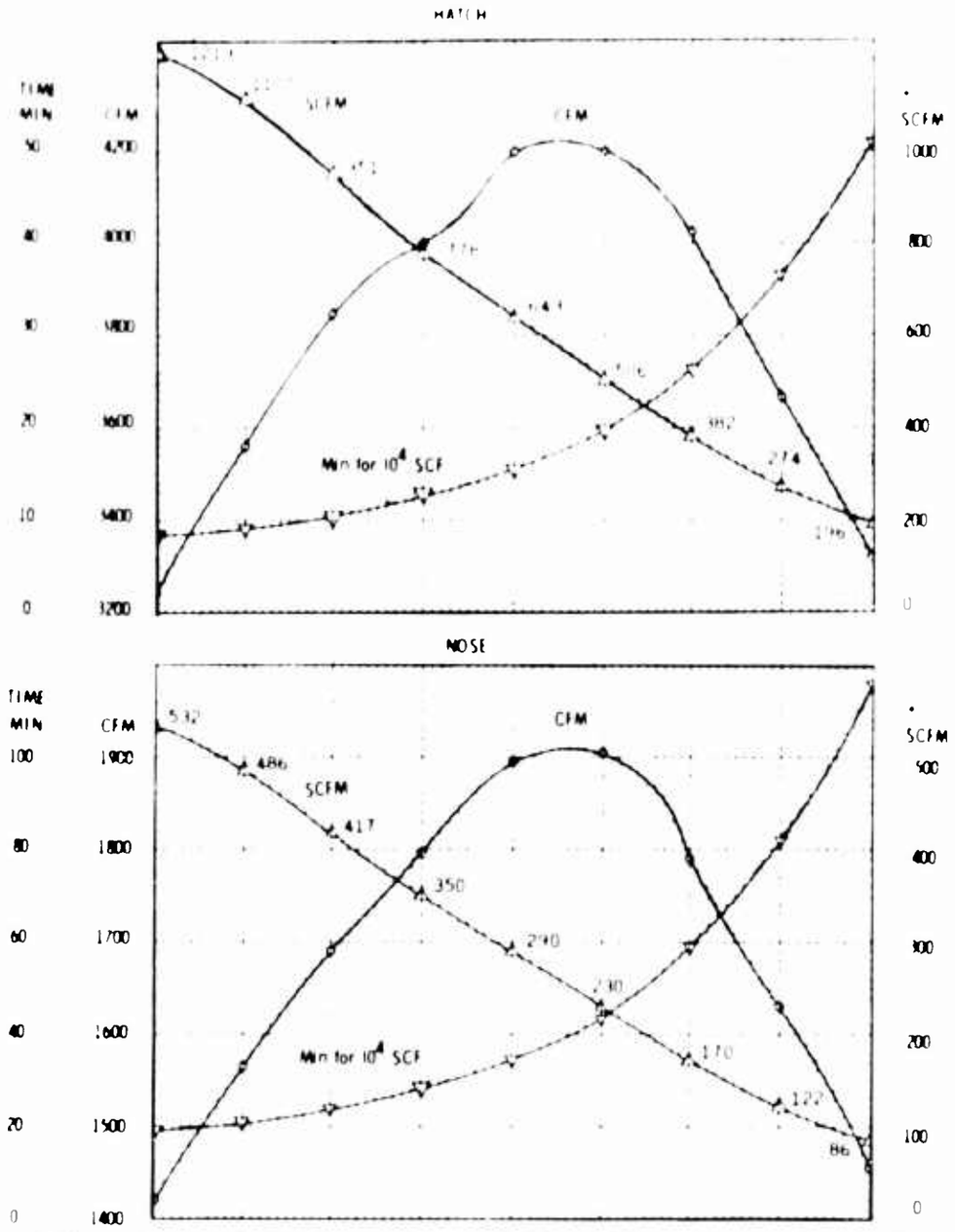
Figure 6 shows the flow rates based upon the Reid data. Figure 7, below, shows the effective change of flow in each of the ducts compared to the flow data previously published in DASA-532.

Notice that the hatch flow rates have been reduced about 8% while the nose flow rates have been reduced from 8% to 18% depending upon altitude. The effect of this change has been to increase the HASP inventories from 10% to 13% over those previously reported. The anlyses which depend upon isotopic ratios are

*See Case 5, Table I, page 15

U-2 DUCT FLOW RATES

Based on E. G. Reids curves Sept 60
and IAS schedule in use 59 60



ACT ALT	30 000	35 000	40 000	45 000	50 000	55 000	60 000	65 000	70 000
IAS CORR	7%	7%	8%	8%	9%	10%	11%	13%	15%
TEMP CORR	4%	4%	5%	5%	5%	5%	6%	6%	7%
IAS	175	173	170	165	160	150	135	121	108
TAS	280	303	340	361	391	410	414	416	420
IND TEMP	54	42	42	39	36	34	34	33	33
MACH NO	0 48	0 53	0 58	0 63	0 68	0 71	0 72	0 73	0 73

FOR EVERY 10 KNOTS IAS HIGH ADD IAS CORR % TO CFM AND SCFM
FOR EVERY 10^{0} COLDER IND TEMP ADD TEMP CORR % TO CFM ONLY

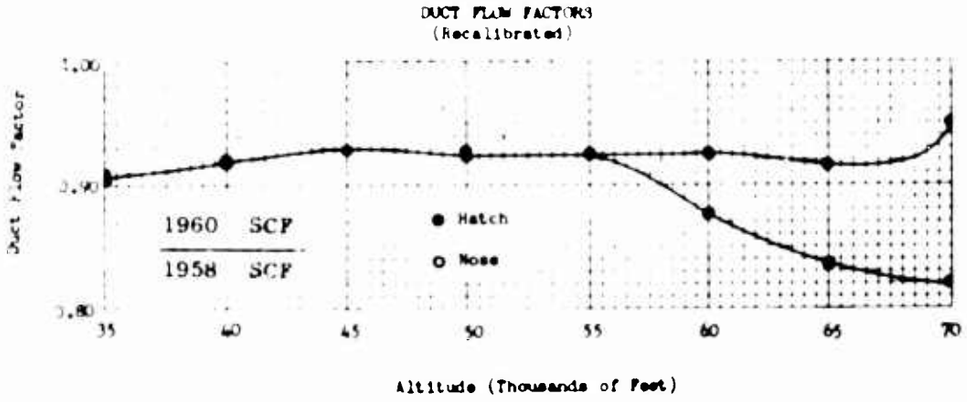
* 760 mm Hg 15°C

FIGURE 6

unaffected if the values for the two isotopes are taken from the same paper. This is the usual practice.

The effect of the variability of different filter papers has been shown[20] to amount to less than 6%. In view of this fact and in light of the foregoing discussion, it is felt that the nose flow rates reported here are good to ±10% and that the hatch flow rates are good to +20%, -0%.

Figure 7



Intercomparison with the B-57 Samplers

During the Air Sampling Program 1960[21,37], conducted from Kirtland Air Force Base, New Mexico, the opportunity presented itself to intercompare the Los Alamos Scientific Laboratory and F-33 air sampling equipment mounted on B-57 aircraft with the nose sampling equipment flown by U-2 aircraft at Minot Air Force Base, North Dakota.

Three LASL-U-2 nose and two LASL-F-33 intercomparison tests were conducted in March 1960 and the data are presented in Table 1. For cases 1 and 2 the B-57 aircraft flew in close proximity to the U-2 and the filter papers were exposed virtually simultaneously. In case 3 a space and time difference existed between

Table I Sampler Intercalibrations

LASL - U-2 Nose Sampler Comparison of Sr-90 Data

Case 1

Date:	March 22, 1960		March 29, 1960		
Altitude:	40,000 feet		40,000 feet		
Aircraft:	B-57	U-2	B-57	B-57	U-2
Filter No:	WB 091	HASP 3461	WB 133	WB 134	HASP 3498
Open Lat.	50°15'N	50°17'N	46°07'N	43°30'N	50°17'N
Long.	102°32'N	102°31'W	100°40'W	100°30'W	102°31'W
Time	1400 Z	1359 Z	1542 Z	1610 Z	1500 Z
Close Lat.	56°21'N	56°21'N	38°45'N	38°45'W	38°01'N
Long.	107°10 W	102°03'W	99°58'W	99°58'W	100°31'W
Time	1523 Z	1525 Z	1700 Z	1700 Z	1716 Z
Sr-90 dpm/1000SCF	43.9±1.3	48.5±0.5	54.7±1.6	53.6±1.6	47.7±0.3
Average:			54.2±1.6		
Difference divided by the mean:	10%				13%

Case 3

Date:	March 29, 1960		
Altitude:	45,000 feet		
Aircraft:	B-57	B-57	U-2
Filter No:	WB 135	WB 136	HASP 3501
Open Lat.	46°50'N	43°45'N	38°17'N
Long.	100°30'W	100°30'W	100°36'W
Time	1920 Z	1950 Z	1928 Z
Close Lat.	43°15'N	40°25'N	50°17'N
Long.	100°30'W	100°15'W	102°31'W
Time	1950 Z	2020 Z	2129 Z
Sr-90 dpm/1000SCF		78.1±2.1	62.0±1.7
Average:	79.2±2.1		
Difference divided by the mean:			24%

Table I (Cont'd)

Case 4. LASL-F-33 Sampler Comparison of Beta Activity (B-57)

Area LASL Sampler Filter Paper: 3.97 ft^2
Area F-33 Sampler Filter Paper: 1.75 ft^2

35,000 ft., Sampling Time 60 min., True Air Speed 375 knots, Flow Rate 1260 SCF/min/ft^2

LASL: Left sampler 787 c/m = 2.62 c/m/1000 SCF
Right sampler 757 c/m = 2.52 c/m/1000 SCF

F-33: Left sampler 423 c/m = 3.20 c/m/1000 SCF
Right sampler 368 c/m = 2.78 c/m/1000 SCF

45,000 ft., Sampling Time 60 min., True Air Speed 320 knots, Flow Rate 610 SCF/min/ft^2

LASL: Left sampler 4495 c/m = 30.9 c/m/1000 SCF
Right sampler 4551 c/m = 31.3 c/m/1000 SCF

F-33: Left sampler 1945 c/m = 30.8 c/m/1000 SCF
Right sampler 2026 c/m = 31.6 c/m/1000 SCF

Case 5: Comparison of Calculated with Measured Relative Collection Efficiencies of U-2 Hatch and Nose Samplers Using 1960 Duct Recalibration

Altitude (feet)	Volume and Activity Ratios of Hatch/Nose				
	Volume	Total β	Sr-90	W-185	Ce-144
70,000	2.28 (29)	2.51 (29)	2.44 (28)	2.43 (15)	2.37 (10)
65,000	2.28 (125)	2.48 (121)	2.44 (122)	2.45 (56)	2.48 (26)
60,000	2.26 (97)	2.50 (90)	2.54 (89)	2.52 (39)	2.58 (14)
55,000	2.20 (30)	2.57 (29)	2.44 (28)	2.54 (6)	2.49 (10)
50,000	2.21 (11)	2.54 (11)	2.50 (38)	2.44 (12)	2.50 (13)
45,000	2.23 (9)	2.41 (9)	2.34 (9)	2.38 (4)	2.55 (3)
40,000	2.26 (3)	2.53 (3)	2.59 (3)	2.58 (2)	3.18 (1)
30,000	2.26 (1)	-----	2.00 (2)	2.80 (1)	------
Average	2.26 (341)	2.50 (325)	2.47 (319)	2.47 (135)	2.50 (77)

The number of samples included in each average are given. It is noted that the Hatch has an apparent collection efficiency which is 10% greater than that of the Nose.

the B-57 and the U-2, and it may not be a valid comparison of sampling equipment. Case 1 shows the results of dual B-57 flights using LASL and F-33 ducts.

Since all samplers used the same type of filter paper (IPC 1178) and the laboratories that performed the isotopic analyses were intercalibrated, it appears that a large part of the differences in the Sr-90 data in cases 1 and 2 are due either to differences in the evaluation of the flow rates in the sampling equipment or to variability of the Sr-90 concentration in the stratosphere. The U-2 data are based on the recalibrated flow rates.

It is difficult to make an accurate appraisal in the absence of more data, but, since the differences in the simultaneous comparisons were relatively small, it can be assumed that the U-2 data can for all practical purposes be integrated with the B-57 data. Further data obtained during Phase V substantiates this conclusion.

Flow Characteristics of IPC 1178 Filter Paper

During the recalibration program two pieces of IPC 1178 filter paper weighing 11.3 grams per ft^2 were tested for their pressure drop-flow rate characteristics. The results are shown in Figure 8. The reduced pressure drop $\sigma' \Delta p/\mu^2$ is plotted against $\sigma v/\mu$ where Δp is the pressure drop in psf, v is the face velocity in ft/sec and σ and μ are, respectively, the relative density and relative viscosity at the face of the filter. An analytical expression which accurately describes this result is:

$$\sigma' \Delta p/\mu^2 = 0.577\ (\sigma v/\mu)^2 + 0.966\ \sigma v/\mu - 0.12 \qquad (1)$$

This expression can also be used to fit the average value curve by Van den Akker previously reported in DASA-532 and DASA-1168 and is an independent check of that work.

Characteristics of IPC 1478
Filter Paper (14.3 g/ft^2)

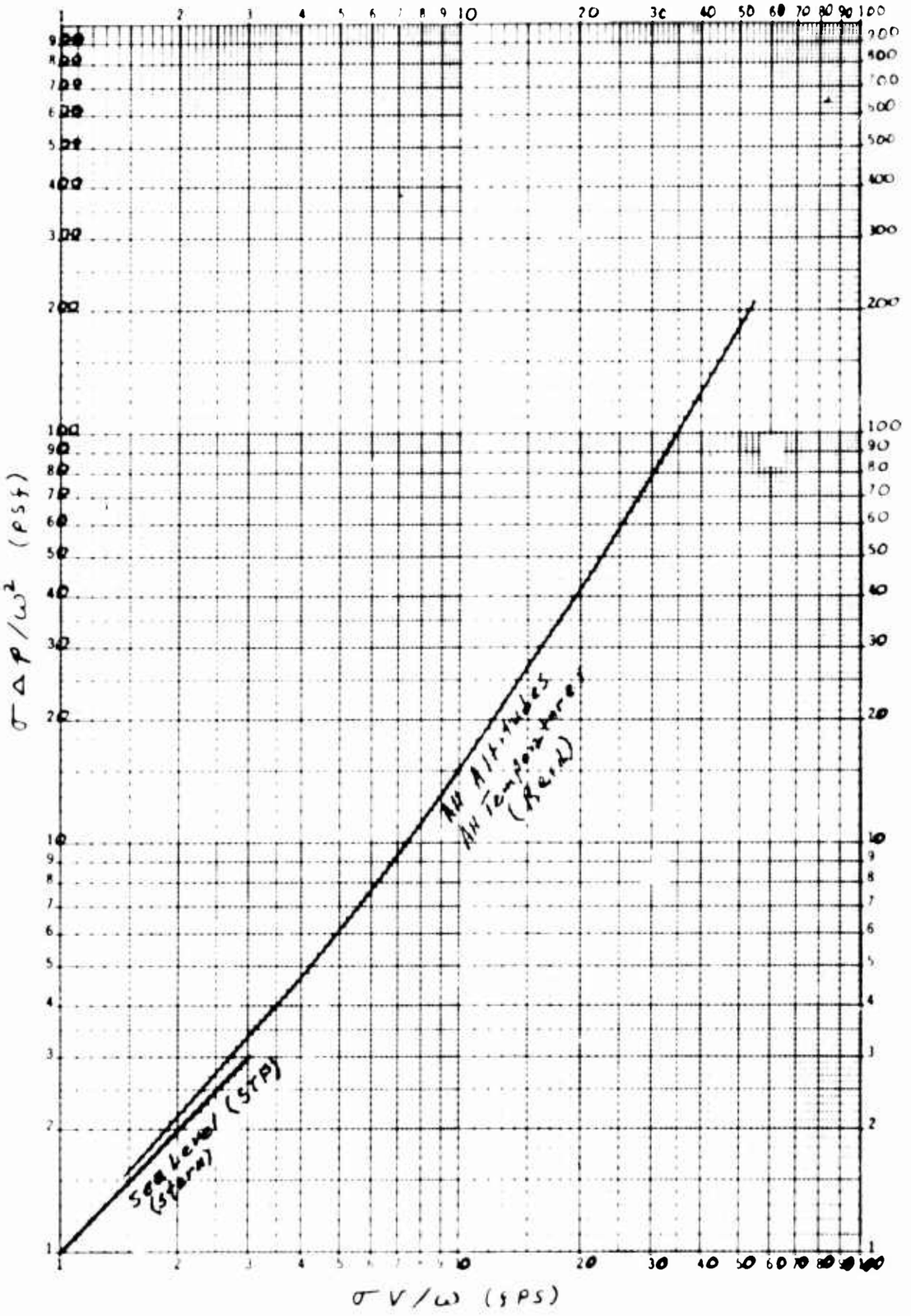


FIGURE 8

Stern has reported filter efficiencies and pressure drop-flow rate characteristics of IPC 1478 filter paper at low flow rates.[23] His generalized expression for flow converted to English units is:

$$v = \Delta p \ (1 + 28.65/p) \qquad (2)$$

where v is the face velocity in ft/sec and Δp and p are, respectively, the pressure drop and face pressure in psf. It can be seen that equations (1) and (2) are not compatible since one is a quadratic and the other is linear and also one is an implicit function of the temperature (μ depends on the absolute temperature alone) while the other is independent of temperature. For sea level conditions (SL on Fig. 8) equation (1) approaches equation (2) as an asymptote and is less than 10% off below Δp equal to 3 psf (1.47 cm H_2O, the highest value of Δp reported by Stern). For conditions at higher altitudes and pressure drops greater than 3 psf equation (2) does not give reliable results, as Stern was careful to point out. Table II shows the results of calculations of face velocity, v^*, using Reid's duct data at various altitudes compared to values of face velocity, v, using Stern's expression extended to higher pressures.

Values as much as 76% different appear when Δp is as high as 76.5 psf. One explanation of this difference is the possibility of compaction of the filter fibers toward the rear of the paper when exposed to a large pressure drop with a subsequent decrease in effective porosity. In addition, flow at high pressures is probably quite turbulent.

The aerosol efficiency reported by Stern[22] confirms the previously reported notion that the filters in the U-2 ducts are very nearly 100% efficient for collecting particles in the 0.1 to 1.0 micron range.

Table II

Comparison of Face Velocities

Altitude	36K	42K	48K	55K	61K	67K	70K
IAS	180	180	160	140	130	120	110
p	570	453	342	256	200	156	132
Δp	76.5	77.1	62.5	49.7	44.5	39.3	33.9
v	80.5	82.0	67.8	55.2	51.0	46.5	41.2
v^*	45.6	49.6	46.6	42.5	41.3	38.2	35.4
v/v^*	1.76	1.65	1.45	1.30	1.23	1.22	1.16

Altitude	36K	42K	48K	55K	61K	67K	70K
IAS	100	100	100	100	100	100	100
p	506	388	297	228	179	142	126
Δp	21.1	24.6	25.0	25.6	26.6	27.8	28.7
v	25.5	26.2	27.1	28.8	30.9	33.1	35.2
v^*	21.1	22.7	24.1	26.1	28.1	29.8	30.9
v/v^*	1.21	1.15	1.16	1.09	1.10	1.12	1.14

v is face velocity according to Stern's method.

v^* is face velocity according to Reid's method.

Chapter III

NATURAL STRATOSPHERIC DUST

Introduction

During the HASP program a subsidiary study to measure the characteristics and distribution of stratospheric particulate matter has been carried on. Originally the effort was aimed at determining the size distribution of the radioactive particles which were being sampled. It was felt that this characteristic of the radioactive debris might play an important part in the sampling efficiency of the IPC 1478 filter paper in use in the sampling ducts. Some of these results were reported previously in DASA-532. Particles in the size range 0.01 to 1.0 microns were encountered but they could not be directly correlated with the sampled radioactivity. Based on a number of considerations, the conclusion was drawn that the filter paper was very nearly 100% efficient in trapping the particulate radioactive debris sampled. This study has lead into an investigation of the naturally occurring stratospheric dust.

Junge has suggested[13] that the "natural" population of the stratosphere consists of three families, one of tropospheric origin in a size range of less than 0.1 micron, one of stratospheric origin in a size range between 0.1 micron and 1.0 micron, and one of extraterrestrial origin in a size range greater than 1.0 micron. Using a balloon borne Aitken counter sensitive in the 0.01 to 0.1 micron region, Junge has shown that the concentration of the "tropospheric" particles diminishes with increasing height above the tropopause to 1/10th the upper tropospheric concentration within 13,000 feet above the tropopause. (See Fig. 9) Vertical structure in the particle concentration profile suggests horizontal layering of clear stratospheric air with the more contaminated tropospheric air and further suggests some type of horizontal mixing between the troposphere and stratosphere in the mid-latitude regions sampled.

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Using a balloon borne General Mills impactor with peak collection efficiency near 0.15 micron, Junge[13] has also shown the existence of a dust layer extending from above the tropopause to around 80,000 feet, with maximum concentrations near 65,000 feet. (See Fig. 10) Using an Electron Microprobe Analyzer, these particles were shown to contain sulfur, probably in the form of ammonium sulfate. Junge suggests that these particles are formed within the layer where they appear by a primary process of oxidation of H_2S and SO_2 introduced by diffusion through the tropopause. The sulfur density in the balloon collections was shown to be about 3×10^{-15} gms/cm^3 compared to 1000×10^{-15} gms/cm^3 of gaseous sulfur components in the high troposphere.

Aircraft Sampling

Since January 1960 two U-2 aircraft used in HASP sampling have had probes installed on a mount near the nose of the aircraft (See Fig. 1 for location of mount). The sampler consists of a removable cartridge housing two retractable impaction surfaces, one of 1.2 cm^2 area and one on the tip of 0.5 cm^2 area (See Fig. 11). The impaction surfaces are extended and withdrawn simultaneously by the pilot after reaching the sampling altitude.

Collection has been at the rate of about one exposure every two weeks for 3 to 7 hours. Alternate samples are analyzed at Isotopes, Inc., and at AFCRL. The results obtained by Friend and Sherwood[10] at Isotopes, Inc., are reported below.

The impaction surfaces consisted of formvar films coated on 200 mesh gold screen. From each sample four 1/8 inch discs were punched for use as electron microscope specimens. Several electron micrographs of each specimen were made in an effort to ensure representativeness for particle size - frequency measurements. Electron micrographs of some typical stratospheric particles are shown in Figure 12.

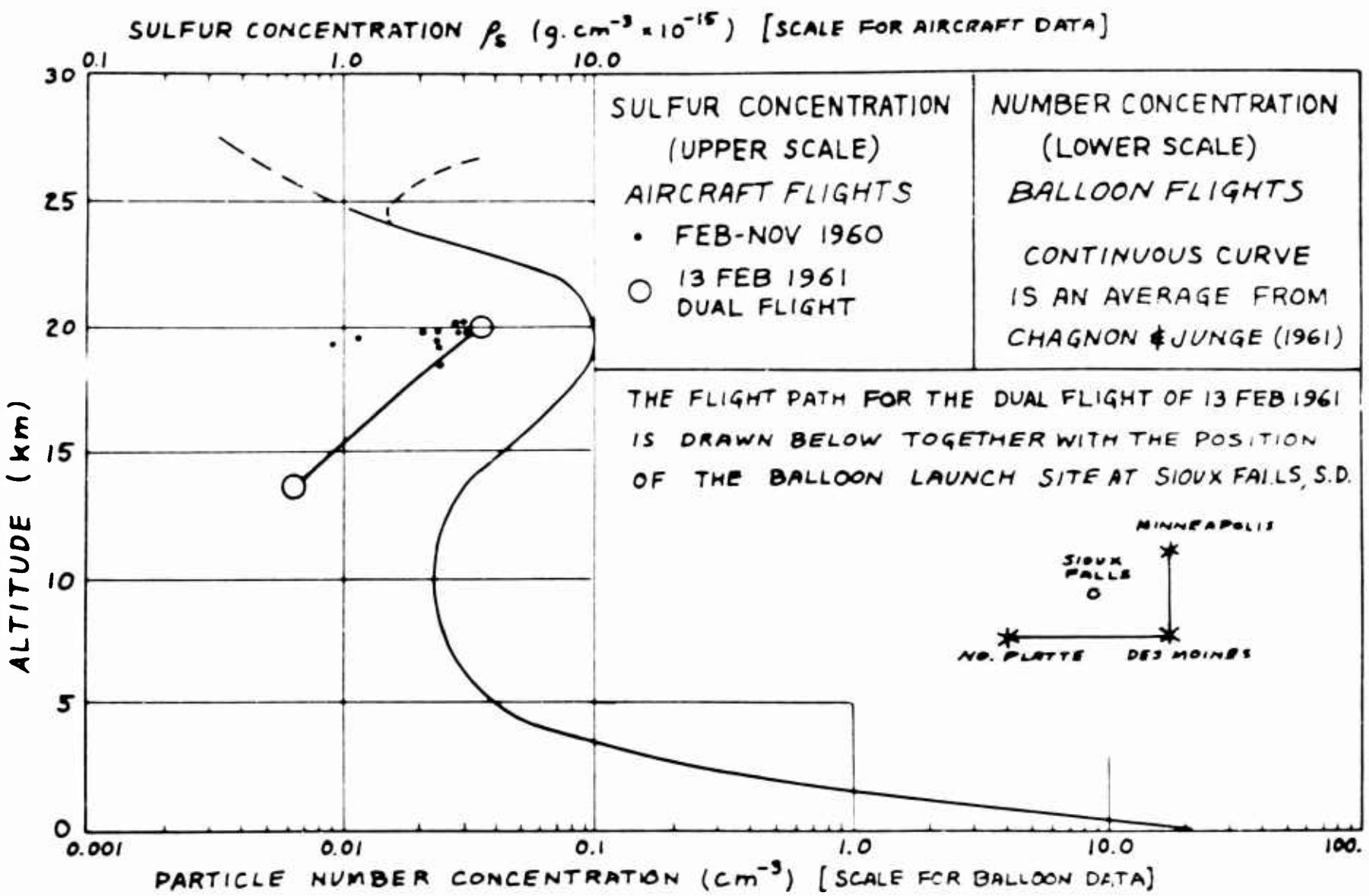


FIGURE 10. Comparison of sulfur density versus altitude and number concentration versus altitude.

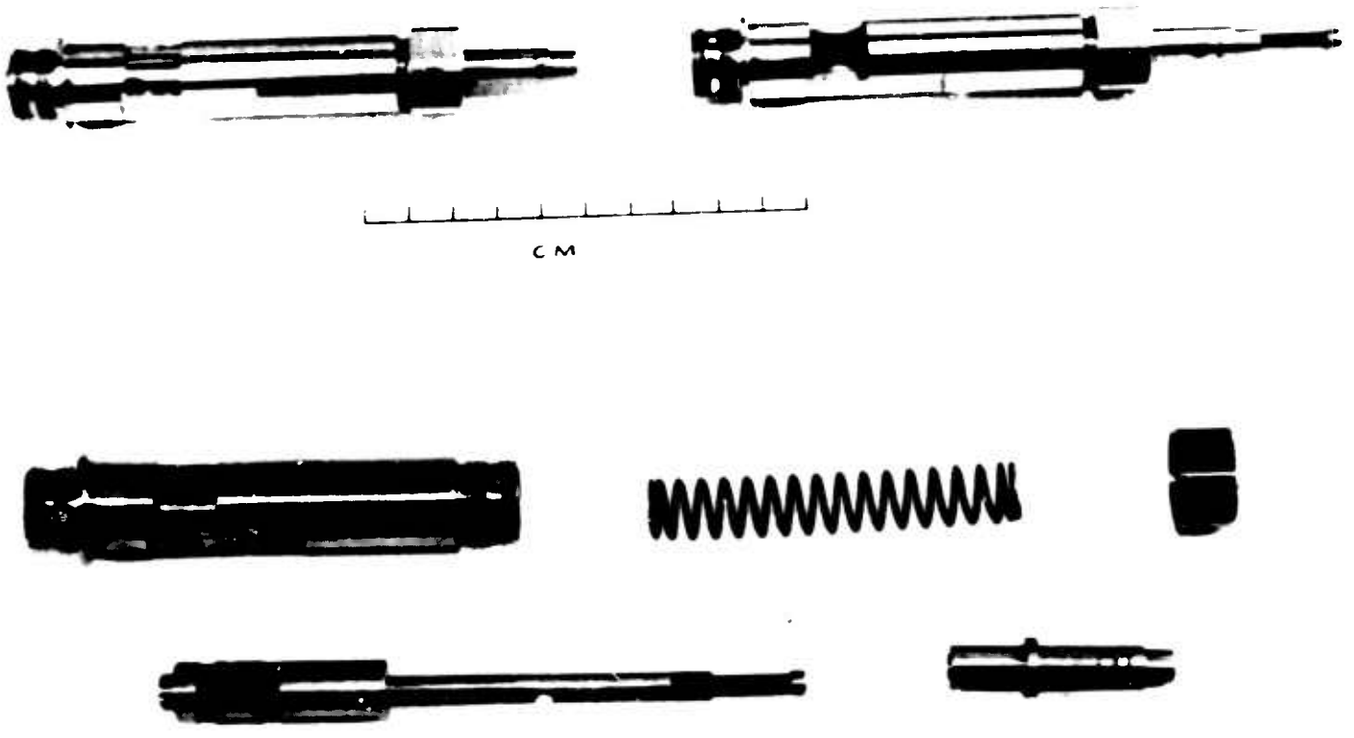


Figure 11 Dust Probe Cartridge

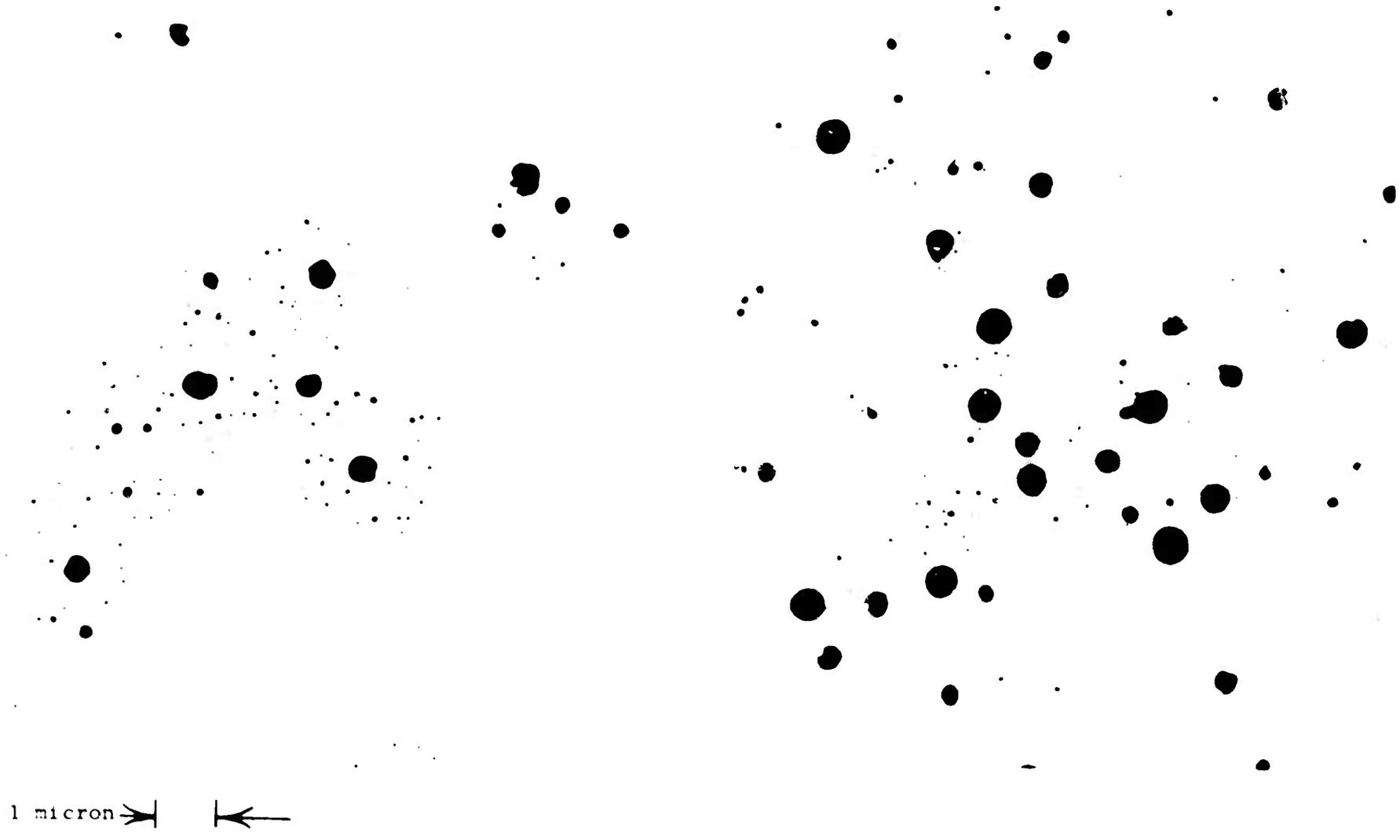


Figure 12 Electron Photomicrographs of Sample W-11 (5000X)

Most of these particles, which have radii in the range from less than 0.10 micron to 1.5 microns, gave electron diffraction patterns corresponding to ammonium persulfate, $(NH_4)_2S_2O_8$. Some of the particles were shown to be ammonium sulfate, $(NH_4)_2\ SO_4$, by measurement of crystal angles and by electron diffraction.

The sulfate and persulfate particles were hygroscopic and were found to be somewhat volatile in the electron beam. The identification of sulfate and persulfate as constitutents of stratospheric particles represents an independent corroboration of the work of Junge and co-workers.

Few of the particles with radii greater than 1.5 microns contain sulfate. Most of these large particles have a higher electron optical density and more irregular outlines than the sulfate particles. Occasionally spheres of high density with diameters smaller than one micron were found. It is possible that some of these nonsulfate particles are extraterrestrial in origin.

The particles as they appeared in the electron micrographs, were counted and classified according to radius. The number of particles in each class was corrected for impaction efficiency for a cylindrical surface according to the method of Ranz and Wong[23] using a particle density of 2 gm/cm^3. The density of $(NH_4)_2\ S_2O_8$ is 1.982, that of $(NH_4)_2SO_4$ is 1.769. Because the impaction efficiency for particles smaller than 0.1 micron radius is very small (<2%) the lower limit of radius of particles classified was 0.1 micron. Figure 13 shows the average distribution of particle radii plotted as $\frac{dn}{d(\log r)}$ per cm^3 (concentration) vs r (radius), where dn is the number of particles per cm^3 of air with radii in the interval d(log r). The dashed portion of the distribution represents an extrapolation from 0.16 to 0.10 micron radius. In all, the results of classification of sixteen samples, covering the altitude range between 50,000 and 70,000 feet, were used to compute the average distribution of particle radii. For altitudes lower than 60,000 feet the collection efficiencies for particles with

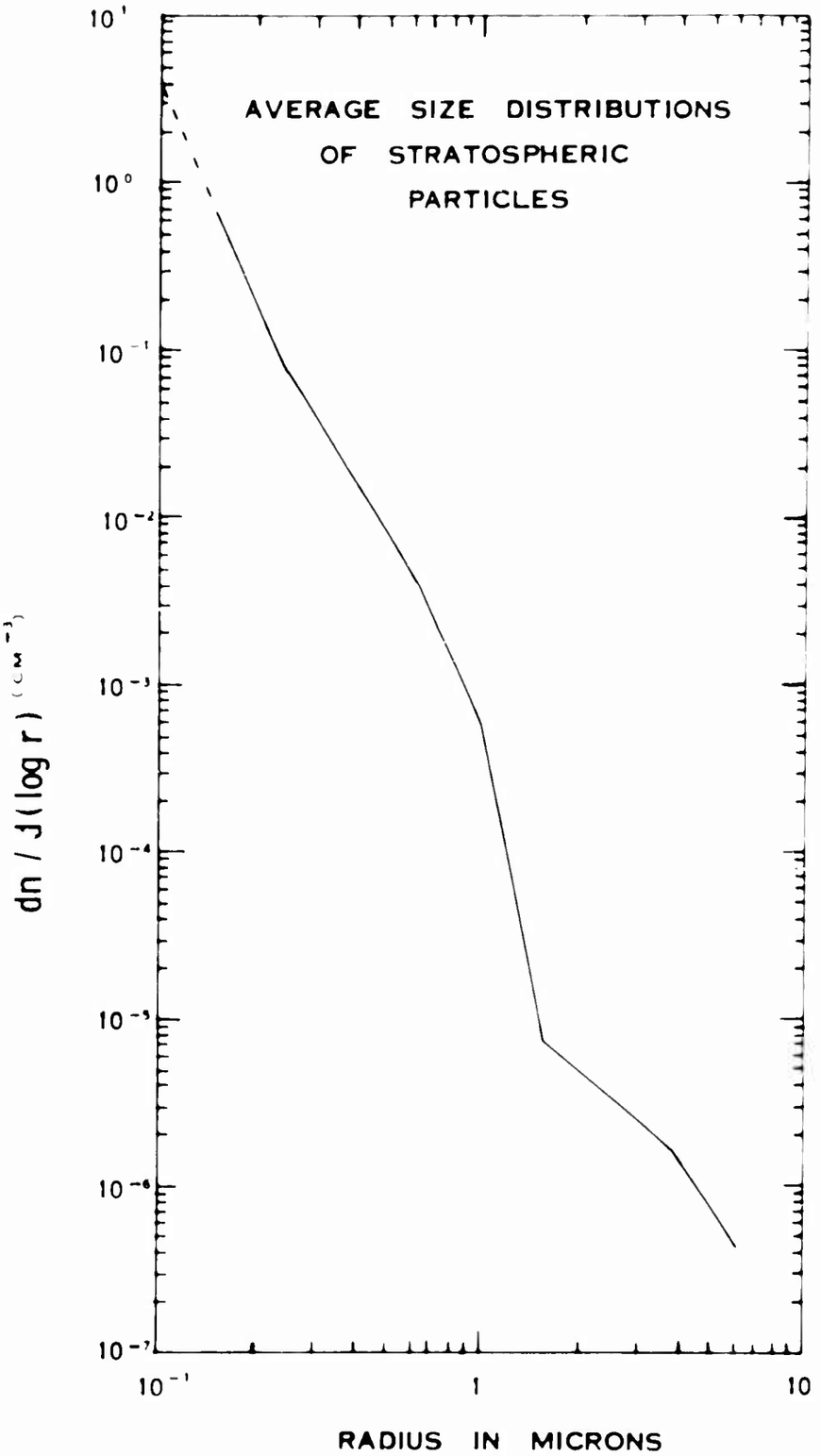

AVERAGE SIZE DISTRIBUTIONS
OF STRATOSPHERIC
PARTICLES
dn / d(log r) (cm⁻³)
RADIUS IN MICRONS
10¹
10⁰
10⁻¹
10⁻²
10⁻³
10⁻⁴
10⁻⁵
10⁻⁶
10⁻⁷
10⁻¹
1
10

FIGURE 13

radii less than one micron are low and strongly dependent upon altitude. For this reason the portion of the distribution curve shown in Figure 13 for radii less than 0.16 micron was determined primarily by the results of nine samples all collected at mean altitudes greater than 61,000 feet. For particles with radii greater than or equal to 0.1 micron all sixteen samples were included in the averaging, there not being a discernible altitude dependence for particles in this size range. The average curve yields a number concentration of 1.1 particles/cm^3 and a volume concentration of 9.6×10^{-15} cm^3/cm^3. The number concentration is determined almost entirely by the number of particles in the smallest size range considered here ($0.079\mu \leq r \leq 0.126\mu$, or $\bar{r} = 0.100\mu$). Although many particles of this size range were observed in the samples, their concentration was determined by extrapolation as previously explained. It is therefore considered that the number concentration may be in error by as much as a factor of five. The volume concentration of particles in the extrapolated region is about 10% of the total calculated from the average distribution. Thus the error in the volume concentration due to the extrapolation is probably not more than about $^{+\ 200\%}_{-\ \ 10\%}$. The volume concentration of particles with radii between one micron and 6.3 microns (the largest particles found in the samples) is about 7×10^{-16} cm^3/cm^3, as computed from the average distribution.

Table III lists data pertinent to the nine samples collected in the 65,000 feet altitude region. The individual number concentrations and volume concentrations listed were calculated using collection efficiencies based on the average distribution and are therefore dependent upon the extrapolation as explained above. Also listed in the table are some strontium-90 concentrations (in dpm/cm^3) of the air sampled by the probes. The strontium-90 concentrations were obtained from HASP filter paper samples which were exposed simultaneously with the particle collections, and are average values over the flight paths. There is indicated a relatively small

Table III Summary of Particle and Radioactivity Concentrations

Sample No.	Date Collected	Latitude	Altitude (feet)	$n^{\dagger}$ (cm^{-3})	v^{*} (cm^3/cm^3)	Sr^{90} Concentration (dpm/cm^3)
W-3	3/16/60	27N⇄12N	67,000	1.57	1.24×10^{-14}	3.61×10^{-7}
W-8	4/5/60	15N⇄8S	67,000	0.94	1.17×10^{-14}	2.70×10^{-7}
W-10	4/21/60	48N	65,000	3.56	3.36×10^{-14}	6.17×10^{-7}
W-12	4/30/60	66N→50N	65,000	1.19	9.43×10^{-15}	5.02×10^{-7}
W-13	5/9/60	15N⇄6S	67,000	1.42	1.01×10^{-14}	3.03×10^{-7}
W-14	5/12/60	16N→29S	65,000	0.86	1.31×10^{-14}	2.84×10^{-7}
W-16	11/17/60	62N→32N	64,000	0.10	4.14×10^{-15}	-----
W-17	11/21/60	15S→20N	65,000	0.63	6.55×10^{-15}	-----

† Number comcentration (based upon extrapolation of particle size-concentration distribution to 0.1 micron radius)

* Volume concentration (~40% contributed by extrapolated portion of particle size-concentration curve)

variation from sample to sample in the ratio of strontium-90 concentration to the volume concentration of particles. The larger ratio exhibited by sample W-12 may be explained in part by the presence of strontium-90 from the high alitude rocket detonations Teak and Orange (See Chapter VIII) in addition to the strontium-90 from the lower stratospheric injections of the HARDTACK test series. Debris from this latter source was probably the main radioactive component of the particles in the other samples.

Two conclusions may be reached on the basis of the particle size concentration spectrum and the particle chemical compositions.

1) There appears to be a relatively persistent aerosol layer in the stratosphere extending to an altitude above 70,000 feet (to about 80,000 feet according to Junge[12]).

2) Particles composed of ammonium persulfate and ammonium sulfate comprise almost all of the aerosol in the radius range from 0.1 - 1.5 micron which, in turn, comprises more than 90% of the total mass of the aerosol.

The most likely source of the sulfate particles is H_2S and SO_2 of terrestrial origin, which enter the stratosphere by some mixing process and are subsequently oxidized to $SO_4^=$ by ozone and ultraviolet radiation[13].

The average distribution of particle radii shown in Figure 13 differs from that found by Junge et al[13] in the region $0.1\,\mu \leq r \leq 1.0\,\mu$ in both slope and intercept at 0.1μ. The value of $dn/d(\log r)$ at 0.1μ obtained by Junge is about a factor of five lower than found in this work. Junge found that $dn/d(\log r)$ varied as $1/r^2$ for this region while in this work $dn/d(\log r)$ varies approximately as $1/r^1$. These discrepancies point up the rather large uncertainties in the determination of the number concentrations of particles with radii smaller than 0.2 micron. However, the volume concentrations represented by the respective distribution functions differ by only a factor of two (this work yielding the lower) in the region 0.1μ to 1.0μ.

By invoking some rather crude assumptions the total sulfate content of the stratosphere may be estimated by two methods which are outlined below:

1) Assuming that the aerosol and the strontium-90 in the stratosphere had mixed throughout the layer in which the aerosol exists (tropopause to 80,000 feet) both laterally and vertically, then the total $SO_4^=$ content of the stratosphere is given by the ratio of the average $SO_4^=$ concentration g/cm^3 at 65,000 feet to the average strontium-90 concentration (dpm/cm^3) at the same altitude multiplied by the total

stratospheric burden of strontium-90. The inventory of lower stratospheric strontium-90 exclusive of that from Teak and Orange was about 0.8 megacurie during the period January to June 1960 (See Chapter V). Less than 10% of that amount was estimated to be above 80,000 feet. The mean concentration of strontium-90 at 65,000 feet during that time was 3.1×10^{-7} dpm/cm^3 (ambient). This is comparable to the strontium-90 concentrations listed in Table III. Assuming the aerosol to be composed of ammonium sulfate, the average $SO_4^=$ concentration corresponding to the distribution in Figure 13 is 1.31×10^{-11} gm/cm^3 expressed as $SO_4^=$. The total $SO_4^=$ burden of the stratosphere is thus calculated to be 8.1×10^7 kilograms.

2) Assuming that the vertical profile of aerosol particles is constant relative to the tropopause over all latitudes and that the aerosol density is proportional to the number concentration, as indicated by Junge's work (see Figure 10) and by the relative constancy of the ratio of volume concentration to number concentration in Table III, then integration of the vertical profile over altitude and around the earth yields the total number of particles in the stratosphere (equal to 3.6×10^{21} particles using an average number concentration of 1 particle/cm^3 at 20 km altitude.) This number multiplied by the ratio of average sulfate concentration to average number concentration (Table III) yields a stratospheric sulfate burden of 4.5×10^7 kilograms. The result of using Junge's particle size-concentration distribution in this calculation is 11×10^7 kilograms of $SO_4^=$.

With the assumptions made in (1) above plus the assumption of a 1.0 year residence half-time for strontium-90 of tropical origin, the stratospheric production rate and removal rate may be estimated to be approximately 5.5×10^7 kilograms of $SO_4^=$ per year based on a sulfate burden of 8×10^7 kilograms.

AFCRL Results

The concentrations of sulfur found on the U-2 probes analyzed by AFCRL are similar to those obtained at Isotopes, Incorporated,[24]* and have an average volume concentration of about 5×10^{-15} cm^3/cm^3. This is equal to 2.3×10^{-15} gm/cm^3 of elemental sulphur or 9×10^{-15} gm/cc expressed as sulfate and corresponds roughly to the value obtained by balloon sampling (see Table IV).

Table IV

AFCRL 1960 SULFUR DATA

Sample No.	Date Collected	Altitude	Mid Latitude	Elemental Sulfur Concentration $gm/cm^3 \times 10^{-15}$
1	26 Apr	20km	21°N	1.1
2	25 Aug	20km	35°N	2.9
3	26 Sep	20km	37°N	2.0
4	21 Oct	20k	5°-	2.7
5	25 Oct	20km	29°S	3.0
6	------	19km	20 .	0.9
7	26 May	18km	38°S	2.5
8	27 Oct	20km	18°N	2.3
9	27 Oct	20km	50°S	3.0
10	1 Nov	19km	71°N	2.3
11	------	20km	40°N	1.8
12	------	20km	32°N	2.3
13	------	20km	32°N	1.1
			Average	2.3

Elemental Sulfur concentration x 1.1 = $(NH_4)_2 SO_4$ concentration.
Elemental Sulfur concentration x 3.5 = $(NH_4)_2 S_2O_8$ concentration.
Elemental Sulfur concentration x 2 ≅ volume concentration in cm^3/cm^3

*See also Junge CE and JE Manson Journal of Geophysical Research 66, 2163, (1961)

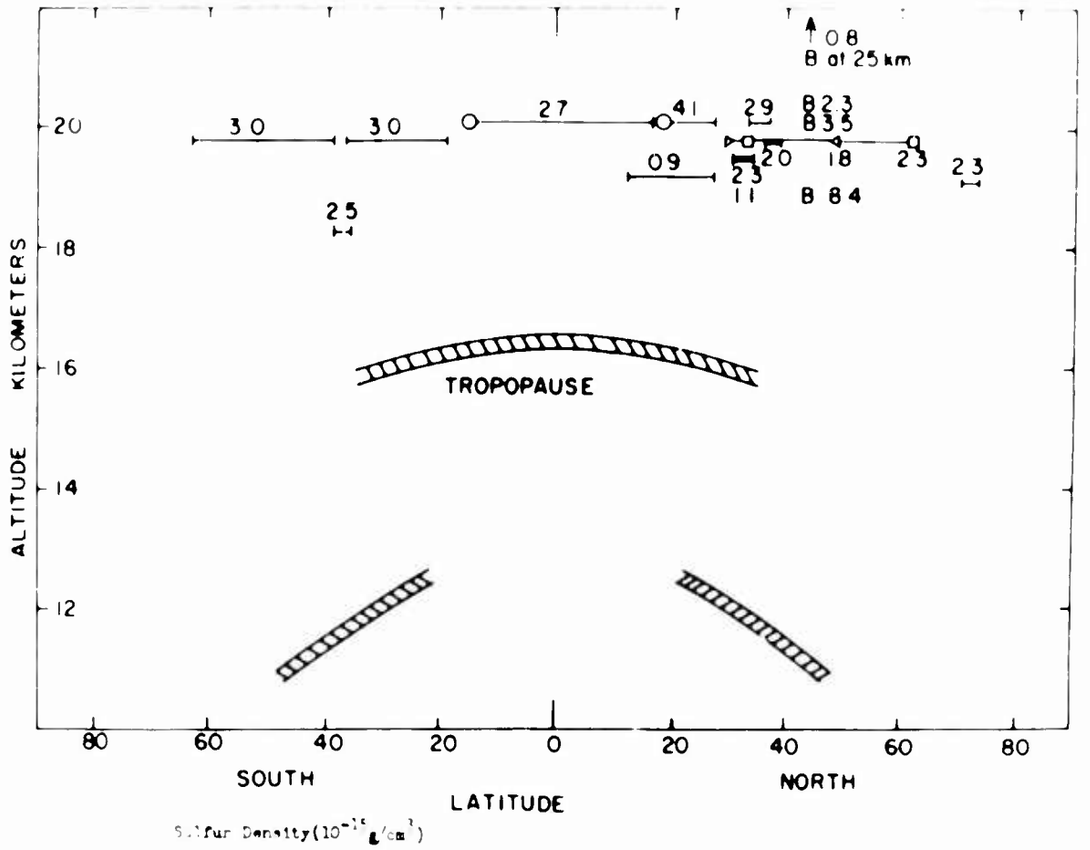
This volume concentration is about half that found by Isotopes, Inc., but is about a factor of four lower than the volume concentration corresponding to Junge's particle radius distribution curve. Figure 11 shows the distribution of sulfur density and particle concentration as a function of altitude and latitude as determined by Junge. These distributions are based on the samples listed in Table IV.

A dual collection was made on 13 February 1961 near 40° N at 65,000 feet and 15,000 feet (20km and 13km). The results showed, respectively, a concentration of 3.6 and 0.7 x 10^{-15} g/cm^3 of elemental sulfur(24)*. These figures are plotted on Figure 10 and show a sulfur density gradient similar to the number concentration gradient for 0.15 micron particles obtained with the General Mills sampler on the balloon.

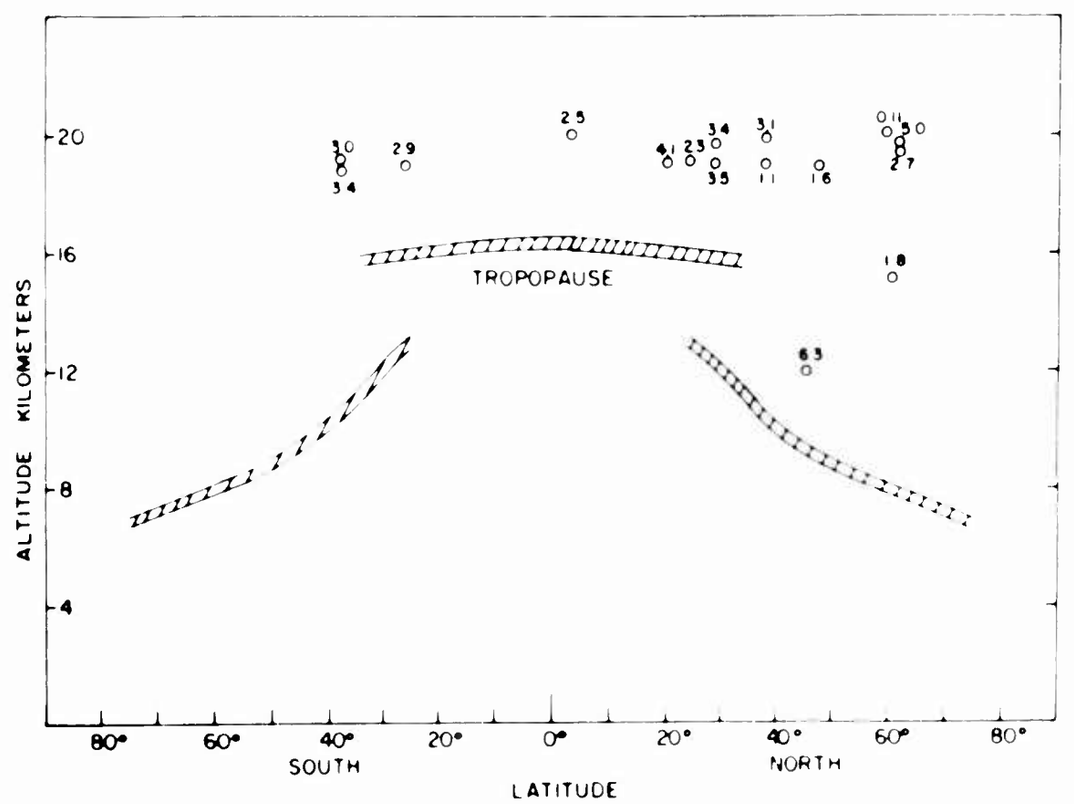
Effects of Dust Layer on Fallout

The aerosol material described above may have a pronounced effect upon the distribution of radioactive debris injected into the lower stratosphere and may have an even more pronounced effect upon measurements of radioisotopic concentrations using various types of sampling equipment. When a radioactive cloud is injected into the stratosphere, the various radioisotopes are spread about in size distributions which may be quite different at different altitudes. Several processes may account for this. For instance, an element like strontium-90 with a rare gas precursor may be in an almost atomic or molecular form as may an element like cesium with a low boiling point, while an element like cerium with a high boiling point may have condensed earlier in the history of the fireball onto unvaporized dirt particles or condensing bomb case materials. The presence of the dust layer would have a tendency to increase

*See also Manson, J. E., C. E. Junge, and C. W. Chagnon "The Possible Role of Gas Reactions in the Formation of the Stratospheric Aerosol Layer" paper presented at Int. Symp. on Chem. Resh. in the Lower and Upper Atmos. San Francisco, Calif, 18 April 1961 (To be published by Interscience)



Sulfur Density(10^{-12} g/cm^3)



Particle Concentrations(0.1 micron to 1.0 micron particles per liter)

Figure 14

the average size of the radioactive particles by contributing to their coagulation into the background material. The rates of coagulation depend upon the initial size of the particles. Junge[13] has calculated the coagulation half-life at 20km of 0.005 micron, 0.01 micron and 0.02 micron particles to be, respectively, 20, 80, and 280 days, the very small particles being incorporated into the dust background in a short time compared to the meteorological residence time.

Particles larger than a few hundredths of a micron will remain in a more or less separate population, intermixed with the natural particles. At altitudes higher than the top of the dust layer, the disparity in average particle size of various elements may persist for longer periods of time since self-coagulation rates are much smaller (although the mean free path is increased).

Sampling equipment such as that used in the Ashcan Project have efficiencies which are quite dependent on particle size due to the slow airflow rates. Using an efficiency correction for 0.03 micron particle size, the Ash Can and HASP data confirm one another at 65,000 feet[2]. Using the same efficiency correction at 95,000 feet could introduce a departure from the "true" values by as much as a factor of 2 and could thereby induce an "apparent" fractionation in the debris or shift the apparent "age" of the debris (See Chapter VI).

Material incorporated into the natural dust layer may depart from the lower stratosphere at a rate different from those particles not incorporated into it, although the difference is probably not too marked, since the rate of fall of the dust layer is necessarily slow when compared to the "airflow" between the stratosphere and troposphere. The fact that the HARDTACK tungsten was removed from the tropical stratosphere faster than the HARDTACK strontium and the fact that tungsten mixing poleward was along surfaces that sloped downward may be partially due to the behavior of the dust layer, however, other processes may better account for this

particular behavior (See Chapter V).

When parcels of the stratospheric dust layer are introduced into the upper troposphere, their hygroscopic nature may enhance their early incorporation into precipitation thereby causing fractionation in the rainfall. Often isotopic ratios measured in rain of material originally in the stratosphere show considerably greater variability than the stratospheric air measurements (See Chapter X).

Sampling of the type described above will continue for the next few years using an improved sampler. Some of the samples previously obtained show evidence of absorption of moisture by the collected material at some time between the exposure and the analysis. An air tight cartridge should eliminate this difficulty. It is hoped that further information concerning the structure of the dust cloud and the role it plays in the distribution of radioactive particles will be forthcoming from this program.

Chapter IV

SYNOPSIS OF PHASE V DATA

Sampling Program

During May and June 1960, a number of particulate collections were made at various locations throughout the world at altitudes ranging from 15,000 feet to 70,000 feet. B-57 and U-2 aircraft were operated out of Argentina, Puerto Rico, Southwest US, Alaska, and the Philippines. This was the first of the semiannual spot check programs described in Chapter I. Southern Hemisphere operation during November 1960 (Crowflight Phase VI) was transferred from Argentina to Australia. Analyses of the samples were performed by several intercalibrated laboratories including Isotopes, Inc. and AFCRL.

Results

The analyses performed by Isotopes, Inc. during Crowflight Phase V were confined to the samples collected by U-2 aircraft and are shown in Table V. Filter papers marked WE were exposed in the nose duct and those marked FD were collected in the hatch duct. The new flow rates given in Chapter II were used to determine the sample volumes. Altitudes are expressed in thousands of feet. All isotopic concentrations are corrected back to date of sampling. The persistent 10% difference between the nose and hatch ducts mentioned in Chapter II is evident in these collections.

Table VI displays additional data obtained during mid and late 1960[25] In this case the U-2 data is from papers exposed in the hatch duct only and is based on the new flow rates given in Chapter II. The B-57 aircraft used the F-33 duct. The two nose filter papers shown in Table V for 23 May 1960 were exposed

Table V

ISOTOPES, INC. DATA FOR CROWFLIGHT PHASE 5

dpm/1000 SCF

Sample No.		Latitude	Altitude	Total Beta	Sr^{90}	W^{185}	Ce^{144}	Rh^{102}	Be^{7}	P^{32}
Missions C64S and A64S (HASP 64C) 9 May 1960										
WE1708	(3802)	15:00N/9:25N	60	2390	55.6 ± 1.1	1.86 ± 0.21	298 ± 5			
WE1709	(3803)	2:25S/8:00S	60	670	17.9 ∓ 1.1	"	93.5 ∓ 3.9			
WE1710	(3804)	9:11N/3:37N	60	1333	25.3 ∓ 0.7	"				
WE1711	(3805)	3:23N/2:11S	60	550	7.60 ∓ 0.13	"				
FD897	(3806)	3:23N/2:11S	60	≤760	15.8 ∓ 0.5					
FD898	(3807)	2:25S/8:00S	60	1180	24.2 ∓ 0.5					
FD899	(3808)	8:00S/2:25S	65	2670	56.6 ∓ 0.6	6.92 ± 0.44	295 ± 5			
FD900	(3809)	2:11S/3:23N	65	3030	93.9 ∓ 2.6	"	370 ∓ 9			
FD901	(3810)	3:37N/9:11N	65	5620	131 ∓ 4	"	509 ± 9			
FD902	(3811)			MALFUNCTION						
WE1716	(3812)	15:00N/9:25N	64.2/65.4	5830	127 ± 1	4.21 ± 0.29	636 ± 6			
WE1717	(3813)	9:11N/3:37N	65.4/66.6	6000	123 ∓ 0.1	"	623 ∓ 7			
WE1718	(3814)	3:23N/2:11S	66.6/67.4	4530	103 ∓ 2	"	513 ∓ 5			
WE1719	(3815)	2:25S/6:41S	67.4/68.4	2150	24.2 ∓ 0.6	"				
FD891	(3816)	3:23N/2:11S	66.6/67.4	4220	99.3 ∓ 7.0		493 ± 6			
FD892	(3817)	2:25S/6:41S	67.4/68.4	3110	82.8 ∓ 1.7					
FD893	(3818)	6:41S/2:25S	68.4/69.1	3410	99.2 ∓ 1.7					
FD894	(3819)	2:11S/3:23N	69.1/69.5	6530	131 ∓ 2	7.17 ± 0.07	694 ± 6			
FD895	(3820)	3:37N/9:11N	69.5/70.3	6110	156 ∓ 3	"	835 ∓ 7			
FD896	(3821)	9:25N/15:00N	70.3/71	9720	205 ∓ 5	"	1298 ∓ 14			
Missions A64R and C64R (HASP 64R) 12 May 1960										
WE1664	(3849)	16:25N/5:08N	64.5/66	4740	111 ± 1	4.26 ± 0.25	561 ± 6			
FD1000	(3853)	16:25N/5:08N	64.5/66	5790	119 ∓ 1	"	664 ∓ 5			
WE1660	(3865)	16:25N/4:50N	63/66	5180	126 ∓ 4	"	736 ∓ 7			
FD904	(3869)	16:25N/4:50N	63/66	6160	133 ∓ 0.05	"	751 ∓ 6			
WE1665	(3850)	4:54N/6:23S	66/68	3410	94.8 ∓ 1.2	8.62 ± 0.30	417 ∓ 5			
FD1001	(3854)	4:54N/6:23S	66/68	3960	108 ∓ 0.1	"	483 ∓ 5			
WE1661	(3866)	4:36N/6:00S	66/67	3040	95.4 ∓ 1.5	"	394 ∓ 7			
FD905	(3870)	4:36N/6:00S	66/67	3350	108 ∓ 0.1	"	470 ∓ 6			
WE1666	(3851)	6:37S/18:36S	68/69	3090	89.6 ∓ 1.5	10.4 ± 0.4	429 ∓ 7			
FD1002	(3855)	6:37S/18:36S	68/69	3710	104 ∓ 1	"	469 ∓ 5			
WE1662	(3867)	6:14S/17:44S	67	3560	98.2 ∓ 0.2	"	446 ∓ 8			
Missions A64R and C64R (HASP 64R) 12 May 1960 (cont.)										
FD906	(3871)	6:14S/17:44S	67	3760	119 ± 0.5	10.4 ± 0.4	445 ± 5			
WE1667	(3852)	18:50S/30:10S	69	4100	98.5 ∓ 2.7	5.46 ∓ 0.33	552 ∓ 6			
FD1003	(3856)	18:50S/30:10S	69	1320	58.0 ∓ 1.4	"	279 ∓ 4			
WE1663	(3868)	17:58S/29:20S	67	4740	137 ∓ 3	"	663 ∓ 7			
FD907	(3872)	17:58S/29:20S	67	5640	136 ∓ 2	"	725 ∓ 6			
Missions E 67 and E 68 and E 69 19 May 1960										
WE1300	(3892)			MALFUNCTION						
WE1302	(3894)	39:45S/36:50S	65	6580	165 ± 3	≤4.23	819 ± 8			
FDE67-2	(3895)	39:45S/36:50S	65	8410	"	"	"			
WE1320	(3896)	36:10S/39:50S	60	3330	117 ± 2	≤4.50	618 ± 5			
WE1322	(3898)	39:45S/36:50S	60	5260	"	"	"			
FDE68-2	(3899)	39:45S/36:50S	60	6530	"	"	"			
WE1332	(3900)	36:40S/39:50S	50	1350	46.0 ± 0.3	3.81 ± 0.04	183 ± 3			
WE1334	(3902)	39:50S/36:45S	50	2100	"	"	"			
FDE69-2	(3903)	39:50S/36:45S	50	2030	"	"				
Missions E 70 and E 71 23 May 1960										
WE1557	(3911)	40:00S/40:00S	64.7/66.7	7100	174 ± 4	[illegible] ± 0.25	871 ± [illegible]	Too Low	842 ± 22	5.64 ± 0.93
WE1349	(3913)	40:00S/40:00S	60	3050	73.9 ∓ 1.7	[illegible] ∓ 0.13	325 ∓ [illegible]	Too Low	539 ± 18	9.97 ∓ 1.99
Missions E 73 and E 74 26 May 1960										
WE1140	(3916)	36:40S/39:50S	60	4220	85.2 ± 1.0	[illegible] ± 2.1	406 ± 5			
WE1142	(3918)	40:00S/36:45S	60	3520	"	"	"			
FDE73-2	(3919)	40:00S/36:45S	60	3600	"	"	"			
WE1204	(3920)	36:40S/39:50S	50	403	10.5 ± 0.4					
WE1205	(3921)	40:00S/40:00S	50	1350	32.0 ∓ 0.4		138 ± [illegible]			
WE1207	(3923)	39:50S/36:45S	50	5960	117 ∓ 4					
FDE74-1	(3924)	40:00S/40:00S	50	1600	15.2 ∓ 0.6					
FDE74-3	(3925)	39:50S/36:45S	65	6480	128 ∓ 2					
Missions E 75 and E 76 and E 77 31 May 1960										
WE1353	(3930)	40:00S/40:00S	64/65	7050	171 ± 1	1.61 ± 0.19	834 ± 3	Too Low	712 ± 20	11.2 ± 1.4
WE1005	(3934)	40:00S/40:00S	60	2880	56.9 ∓ 0.8	3.21 ∓ 0.17	288 ∓ 1		452 ± 14	5.72 ∓ 0.25
WE1137	(3938)	40:00S/40:00S	50	134	3.76 ∓ 0.06					
Missions E 78 and E 79 and XBE 80 2 June 1960										
WE1	(3943)	36:15S/39:50S	61.5/65.5	2250	129 ± 2	≤8.64	639 ± 9			
WE12	(3952)	39:45S/36:50S	66	7340	"	"	"			
WE3	(3945)	39:45S/36:50S	60	1050	47.1 ± 0.2	≤1.81	240 ± 3			
FDE78-2	(3946)	39:45S/36:50S	60	2010	"	"	"			
WE9	(3950)	36:10S/39:50S	60	3790	"	"	"			
WE5	(3947)	36:18S/39:80S	50	726	26.0 ± 0.03	≤2.96	105 ± 2			
WE7	(3949)	39:45S/36:50S	50	1460	"	"	"			
Mission E 99 6 June 1960										
WE13	(3953)	42:30S/50:15S	65.6/66.3	7700	164 ± 0.5	≤1.98	974 ± 8			
WE14	(3954)	50:29S/58:00S	66.3/67.6	6990	180 ∓ 3	"	931 ∓ 7			
WE15	(3955)	58:00S/50:15S	67.6/68.2	8340	202 ∓ 2	≤1.62	1143 ∓ 10			
WE16	(3956)	50:00S/42:30S	68.2/70	10500	227 ∓ 1	"	1259 ∓ 14			
Missions E 91 and E 92 8 June 1960										
WE21	(3974)	31:25S/22:12S	64.5/65.5	4910	121 ± 3	2.51 ± 0.15	581 ± 8		555 ± 8	
FDE91-1	(3978)	31:25S/22:12S	64.5/65.5	5540	134 ∓ 2	"	654 ∓ 4		"	
WE25	(3982)	31:25S/21:37S	63.5/65.4	4950	137 ∓ 3	"	673 ∓ 10		"	
FDE92-1	(3986)	31:25S/21:37S	63.5/65.4	6310	159 ∓ 1	"	683 ∓ 5		"	
WE22	(3975)	21:53S/11:23S	65.5/67	3400	90.3 ∓ 0.6	3.63 ± 0.21	448 ∓ 4		346 ± 7	
FDE91-2	(3979)	21:53S/11:23S	65.5/67	4650	122 ∓ 2	"	538 ∓ 3		"	
WE26	(3983)	21:23S/10:34S	65.4/66	3570	96.9 ∓ 2.4	"	393 ∓ 5		"	
FDE92-2	(3987)	21:23S/10:34S	65.4/66	4200	103 ∓ 2	"	443 ∓ 3		"	
WE23	(3976)	11:08S/01:06N	67/68	3510	93.1 ∓ 1.0	5.31 ± 0.19	373 ∓ 8		221 ± 6	
FDE91-3	(3980)	11:08S/01:06N	67/68	3940	90.0 ∓ 0.6	"	360 ∓ 4		"	
WE27	(3984)	10:21S/00:42N	66.5/65.5	3420	97.3 ∓ 0.8	"	370 ∓ 7		"	
FDE92-3	(3988)	10:21S/00:42N	66.5/68.5	5130	107 ∓ 1	"	437 ∓ 5		"	
WE24	(3977)	01:14N/11:17N	68/69.5	4050	96.0 ∓ 0.6	5.44 ± 0.28	458 ∓ 6			
FDE91-4	(3981)	01:14N/11:17N	68/69.5	4710	127 ∓ 0.1	"	522 ∓ 6			
WE28	(3985)	00:55N/10:48N	68.5/69.7	4360	106 ∓ 1	"	474 ∓ 8			
FDE92-4	(3989)	00:55N/10:48N	68.5/69.7	4870	125 ∓ 2	"	539 ∓ 6			
Missions E 95 and E 93 and E 94 10 June 1960										
WE33	(3960)	19:35N/29:20N	55	1350	27.6 ± 0.2		136 ± 1	1.72 ± 0.19	276 ± 7	
WE29	(3962)	19:50N/29:20N	60	4060	90.9 ∓ 0.2	1.26 ± 0.24	446 ∓ 2	3.11 ∓ 0.36	243 ∓ 8	7.09 ± 0.68
WE31	(3964)	20:40N/29:20N	65	8240	154 ∓ 0.02	2.65 ∓ 0.38	816 ∓ 2	6.92 ∓ 0.79	549 ± 14	7.80 ∓ 0.40

simultaneously on the same aircraft with the corresponding two hatch filter papers shown in Table VI. All the isotopic concentration in Table VI are corrected back to the date of sampling with the exception of the rhodium and tungsten values which are extrapolated to 12 August 1958 and 15 August 1958, respectively. Values for the half lives used were Rh-102, 210 days; W-181, 120 days; and W-185, 76.2 days. No Sr-89 or Ba-140 was found in any of the filter papers. All values shown in Table VI have radiochemical precision with less than 3% error except those marked with the letters a through h which stand for the following values of precision: (a) 3-5%, (b) 5-10%, (c) 10-15%, (d) 15-20%, (e) 20-25%, (f) 25-30%, (g) 30-35%, (h) 35-40%. In general the data in Tables V and VI are compatible when the 10% Nose-Hatch discrepancy is taken into account.

Discussion

Before launching into a detailed discussion of these and other data, it might be worthwhile to summarize here the salient features of the material described above.

Figure 15 shows the strontium-90 concentrations for the periods of May-June 1959 and May-June 1960. The equatorial maximum of 1959 above the tropopause has been replaced by a minimum in 1960 and the maximum values may now be found in the highest latitudes at the highest sampling altitudes. As discussed later in Chapters V and VIII, this is explained by the incursion of debris from the Teak and Orange high altitude shots in the 1958 HARDTACK series. The distribution is much more symmetrical between the Northern and Southern Hemispheres in 1960 than it was in 1959.

Figure 16 displays the tungsten-185 concentrations in the same way that strontium-90 is shown in Figure 15. There is a marked difference in the manner

TABLE VI

ATMOSPHERIC RADIOACTIVITY DATA, MAY-JUNE 1960

Date 1960	May 17	May 20	May 24	May 20	May 24	June 1	May 20	May 24	May 19	May 23
Latitude	70°N	70°N	70°N	34-36°N	35-36°N	36°N	10°N	10°N	40°S	40°S
Longitude	154°W	154°W	154°W	105°W	105°W	105°W	122°E	122°E	62°W	62°W
Altitude (Ft.)	15,000	15,000	15,000	15,000	15,000	15,000	15,000	15,000	15,000	15,000
Aircraft Type	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57
Exposure (Minutes)	120	120	120	120	120	120	120	120	120	120
Volume (1000 SCF)	294	262	286	542	538	523	267	260	378	362
Tropopause (Alt., Ft.)	33,600	30,800	36,600	38,500	41,500	38,600	60,000	56,000	36,000	32,000
Isotope										
Be-7	12.5a	29.7a	7.94a	12.6	9.73	12.6a				
Sr-90	0.775a	1.65	0.767	1.28b		0.896	0.143b	0.073a	0.367	0.172a
Zr-95	0.105	0.188b	0.100b	0.105	0.053a	0.073a				
Rh-102	0.561a	NR	NR	1.20a	NR	NR				
Cs-137	1.50	3.13	1.33	2.03	0.800	0.906	0.236	0.114a	0.588	0.326
Ce-144	4.31	7.50	3.87	5.78	2.17	3.87		0.293		0.794
W-181	16.2	36.3	NR	20.1	NR	14.3a				
W-185	21.1b	NR	25.1a	26.0a	NR	16.9a				
Pb-210	0.785		0.264	0.276	1.16					

Date 1960	May 17	May 20	May 24	May 20	May 24	June 1	May 20	May 24	May 19	May 23
Latitude	70°N	70°N	70°N	33-36°N	35-36°N	36°N	10°N	10°N	40°S	40°S
Longitude	154°W	155°W	154°W	105°W	105°W	105°W	122°E	122°E	62°W	64°W
Altitude (Ft.)	25,000	25,000	25,000	25,000	25,000	25,000	25,000	25,000	25,000	25,000
Aircraft Type	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57
Exposure (Minutes)	120	120	120	120	120	120	120	121	120	120
Volume (1000 SCF)	279	267	292	428	428	437	237	238	405	351
Tropopause (Alt., Ft.)	33,000	30,800	36,600	38,500	41,500	38,600	60,000	56,000	36,000	32,000
Isotope										
Be-7	19.1a	26.1a	8.65b	6.24a	10.7	NR		11.5a	42.5b	
Sr-90	1.89a	1.65	0.814	0.357	0.321	1.08	0.032a	0.495	1.21a	NR
Zr-95	0.204b	0.196a	0.090a	NR	NR	0.090a		NR	0.090a	
Rh-102	NR	NR	NR	NR	NR	NR		NR	NR	
Cs-137	3.67	2.31	1.31	0.658	0.560	1.95	0.053a	0.860	Lost	0.373
Ce-144	9.62	7.79	3.95	1.64	1.49	5.31		2.15	4.76	0.830
W-181	33.1	31.6a	NR	NR	NR	17.5a		NR	37.2a	
W-185	47.9	NR	24.3a	9.57	NR	20.5c		NR	52.6a	
Pb-210			0.275	0.262	0.479			0.641	0.134a	

Date 1960	May 17	May 20	May 24	May 20	May 24	June 1	May 20	May 24	May 19	May 23	June 2
Latitude	70°N	70°N	72°N	34-36°N	35-36°N	35-36°N	10°N	10°N	40°S	40°S	40°S
Longitude	154°W	154°W	154°W	105°W	105°W	105°W	122°E	122°E	62°W	62°W	62°W
Altitude (Ft.)	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000
Aircraft Type	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57
Exposure (Minutes)	120	120	190	120	120	120	120	120	120	120	120
Volume (1000SCF)	229	214	222	229	229	229	190	189	214	189	197
Tropopause (Alt., Ft.)	33,600	30,800	36,600	38,500	41,500	38,600	60,000	56,000	36,000	32,000	44,000
Isotope											
Be-7	339a	239	153a	90.1	154	248a			370	6..4	205a
Sr-90	47.9a	23.0	10.6	5.02	14.6	25.2	Lost	0.057a	15.1a	1.72	3.86
Zr-95	4.76a	2.15b	0.882a	0.644	1.06	1.29a			1.24b	0.147b	NR
Rh-102	16.0a	4.93a	3.64b	NR	3.55a	NR			NR	NR	NR
Cs-137	74.5a	42.2	18.3	9.33	25.6	34.1	0.294a	0.098a	28.0	3.02	6.83
Ce-144	242	125	51.2	24.6	74.7	133		0.263	66.0	7.03	18.6
W-181	541a	299a	129	67.7a	166a	320a			667	64.7b	127b
W-185	784	447a	178a	103a	235a	399a			831a	NR	157a
Pb-210	0.324		0.660	0.655	0.599a				0.350	0.251	

TABLE VI (Cont'd)

ATMOSPHERIC RADIOACTIVITY DATA, MAY-JUNE 1960

Date 1960	May 18	May 20	May 25	May 20	May 24	June 1	May 22	May 24	June 1	May 19	May 23	June 2
Latitude	70°N	70-73°N	70°N	34°N	32°N	32°N	5-10°N	5-10°N	10-15°N	40°S	40°S	40°S
Longitude	157°W	157°W	157°W	105°W	105°W	105°W	34°W	34°W	62°W	63°W	64°W	63°W
Altitude (Ft.)	50,000	50,000	50,000	50,000	50,000	50,000	50,000	50,000	50,000	50,000	50,000	50,000
Aircraft Type	U-2	U-2	U-2	B-57	U-2	U-2	U-2	U-2	U-2	U-2	B-57	U-2
Exposure (Minutes)	287	318	315	120	376	325	232	226	314	352	120	336
Volume (1000SCF)	173	271	236	130	302	251	108	104	145	196	122	204
Tropopause (Mt., Ft.)	33,600	30,800	36,900	38,500	44,000	41,000	52,000	53,000	53,000	32,000	32,000	44,000
Isotope												
Be-7	370a	455	397	161	167	398			1056	3.6a	18.	685b
Sr-90	67.5	76.6	70.3	14.7	13.7	58.8	0.246b	0.364	5.55	48.1a	7.77	31.8
Zr-95	7.15b	3.30a	5.09	1.36a	1.26	4.80a			0.510b	.58a	0.93a	2.22a
Rh-102	28.1a	33.6b	51.0	5.22	7.49a	16.0b			NR	2.70a	NR	3.83a
Cs-137	119	116	121	24.9	23.8	107	0.385a	0.468	7.99	44.1	14.5	51.8
Ce-144	379	402	405	77.3	77.5	326		1.16	27.5	121	40.8	135
W-181	609	705a	482	182	171a	644			1356	674	275	822a
W-185	719b	1226	NR	279b	227	NR			NR	613	NR	1039a
Pb-210	0.319a		0.259	0.476a	0.367					0.204	0.108a	

Date 1960	May 18	May 20	May 25	May 20	May 24	June 1	May 21	May 24	May 31	May 19	May 23	June 2
Latitude	70-73°N	70°N	70-73°N	32°N	32°N	32°N	5-10°N	10°N	7-12°N	40°S	40°S	40°S
Longitude	157°W	157°W	157°W	105°W	105°W	105°W	34°W	34°W	60°W	63°W	63°W	63°W
Altitude (Ft.)	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000
Aircraft Type	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2
Exposure (Minutes)	395	364	406	427	402	256	257	272	368	352	380	318
Volume (1000 SCF)	171	158	177	186	175	111	111	118	160	205	165	138
Tropopause (Mt., Ft.)	33,600	30,800	36,900	38,000	44,000	41,000	53,000	53,000	53,000	32,000	32,000	44,000
Isotope												
Be-7	648	764a	695b	391	485	420b	162	121	265	585	693	385a
Sr-90	161a	171	171a	102	88.6	83.1	17.4	15.5	33.1	100	67.3	63.8
Zr-95	15.3	19.0a	11.1	8.60	6.38b	6.73a	1.66	1.73a	2.28a	10.1a	6.74	4.67a
Rh-102	171	276	223	76.4a	64.3	57.1a	NR	NR	7.62b	75.9a	29.1a	22.7b
Cs-137	288	270	276	177	164	136	29.4	21.3	NR	173	127	104
Ce-144	1113	1087	1314	636	525	445	78.3	93.3	128	529	364	271
W-181	822a	913a	612	798a	646	553a	512	NR	538a	888	1104	857b
W-185	1132	1032a	NR	1086	589a	679a	699	NR	NR	1226	1319	1052a
Pb-210	0.225		0.208a	0.320a	0.439a		0.343	0.273a		0.241a	0.314	

Date 1960	May 18	May 20	May 25	May 20	May 24	June 1	May 21	May 23	June 1	May 19	May 23	June 2
Latitude	70°N	70°N	70°N	32°N	32°N	32°N	5-10°N	5-10°N	10-15°N	50°S	40°S	40°S
Longitude	157°W	157°W	157°W	105°W	105°W	105°W	34°W	34°W	62°W	63°W	63°W	63°W
Altitude (Ft.)	61,500-64,000	62,000-64,400	63,000-65,500	63,000-66,000	64,000-68,700	64,000-66,500	66,500-69,000	64,000-67,000	63,200-64,700	63,500-65,000	64,500-66,500	70,000
Aircraft Type	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2
Exposure (Minutes)	404	403	398	430	443	423	236	255	372	193	354	360
Volume (1000 SCF)	139	136	123	133	126	124	56.9	76.1	119	6	103	73.1
Tropopause (Mt., Ft.)	34,600	30,800	36,900	38,000	44,000	41,000	54,000	53,000	53,000	32,000	32,000	44,000
Isotope												
Be-7	962	1184a	981	563	786b	885a	298b	368c	273b	1037	866	1900
Sr-90	175	185	173	162	180	187	101	72.7	68.1	213	175	229
Zr-95	18.5	26.7a	14.3	15.2	6.68a	15.3	5.31b	6.67a	3.83b	NR	17.0a	19.1a
Rh-102	366	443	331b	118a	209a	126a	25.9	18.0b	NR	164a	176	125b
Cs-137	263a	317	287a	282	311	296	177	119	132	363	293	349
Ce-144	1480	1339	1361	1007	1244	1013	486	362	336	1177	910	1113
W-181	301a	418a	315	715a	496	606a	1780	1613a	1065	1192	1219	1511a
W-185	421	522a	557a	992	783a	886b	2418	1998a	NR	1685	1519a	1845b
Pb-210	0.139a		0.136a	0.266a	0.231a		0.475a	0.568a		0.424a	0.361a	

TABLE VI (Cont'd)

ATMOSPHERIC RADIOACTIVITY DATA, NOVEMBER 1960

Date 1960	Nov 1	Nov 10		Nov 1	Nov 10		Nov 18	Nov 20	Nov 2	Nov 10	Nov 15
Latitude	70°N	71°N		35°N	36°N		9°N	8°N	42°S	40°S	40°S
Longitude	144°W	144°W		105°W	105°W		@122°E	122°E	147°E	147°E	147°E
Altitude (Ft.)	15,000	15,000		15,000	15,000		15,300	15,000	15,000	15,000	15,000
Aircraft Type	B-57	B-57		B-57	B-57		B-57	B-57	B-57	B-57	B-57
Exposure (Minutes)	120	120		120	120		120	120	120	120	120
Volume (1000 SCF)	386	364		521	559		302	302	197	433	433
Tropopause (Kt., Ft.)	34,100	32,300		45,600	37,300		50,000	50,500	34,000	39,000	33,000
Isotope											
Be-	26.9a	13.7a		12.8	8.94b		2.68	2.24c	16.8a	17.7	19.0
Sr-90	0.678	0.272a		0.164a	0.104a		0.023a	0.015c	0.459a	0.369a	0.651
Zr-95											
Rh-103	0.545b	0.236a		0.118a	0.111c		0.067d	0.025d	0.307c	0.289a	NR
Cs-137	1.10	NR		0.286	0.175			0.026a	0.782	0.566	0.955
Ce-144	2.36	1.03		0.595	0.394		0.085b	0.062c	1.35	1.15	1.81a
W-181	7.19d										9.66
W-185	17.1a										NR
Pb-210	0.713	0.940a		0.629	0.447a		0.050a	0.030	0.405	0.111a	

Date 1960	Nov 1	Nov 10	Nov 16	Nov 1	Nov 10		Nov 18	Nov 20	Nov .	Nov 10	Nov 15
Latitude	70°N	71°N	71°N	35°N	36°N		8°N	8°N	4.°S	41°S	40°S
Longitude	144°W	144°W	144°W	105°W	105°W		122°E	122°E	147°E	147°E	147°E
Altitude (Ft.)	25,000	25,000	25,000	25,000	25,000		25,000	25,000	25,000	25,000	25,000
Aircraft Type	B-57	B-57	B-57	B-57	B-57		B-57	B-57	B-57	B-57	B-57
Exposure (Minutes)	120	121	120	120	120		120	120	120	--	120
Volume (1000 SCF)	336	301	323	405	443		231	231	310	0(	298
Tropopause (Kt., Ft.)	34,100	32,300	27,200	45,600	37,300		50,000	50,500	34,000	39,000	33,000
Isotope											
Be-7	53.5	34.5a	108	20.7a	12.9a		2.68	2.35	22.5a	2..3a	26.7
Sr-90	1.35a	0.757	0.811	0.041a	0.085b		0.049b	0.011c	0.636a	0.561a	0.668
Zr-95		NR									
Rh-103	0.664a	0.570b	2.08	0.050a	0.058c		0.191d	0.066d	0.309b	0.195b	NR
Cs-137	2.30	1.22	1.25	0.072	0.163		0.084	0.021a	1.04	0.870	1.12a
Ce-144	4.65	2.84	3.30a	0.121a	0.336		0.229	NR	2.00	1.55	2.02
W-181	13.3a	24.6a	14.0d						15.4d		12.8
W-185	29.2a	47.9a	NR						19.0b		NR
Pb-210	0.378	0.486		0.324	0.269		0.032a	0.035	0.263	0.424a	

Date 1960	Nov 1	Nov 10	Nov 16	Nov 1	Nov 10	Nov 15	Nov 18	Nov 20	Nov 2	Nov 10	Nov 15
Latitude	70°N	71°N	71°N	35°N	36°N	38°N	8°N	8°N	42°S	41°S	41°S
Longitude	144°W	144°W	144°W	105°W	105°W	105°W	122°E	122°E	147°E	147°E	147°E
Altitude (Ft.)	40,000	40,000	40,000	40,000	40,000	40,000	40,000	40,000	40,000	40,000	40,000
Aircraft Type	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57	B-57
Exposure (Minutes)	122	120	120	120	120	120	120	125	120	120	120
Volume (1000 SCF)	193	316	190	265	264	264	164	171	267	229	205
Tropopause (Kt., Ft.)	34,100	32,300	27,200	45,600	37,300	36,700	50,000	50,500	34,000	39,000	33,000
Isotope											
Be-7	361	437	321	21.0b	187	56.1	3.85c	2.65b	103a	248	505
Sr-90	18.0	26.2	14.0a	0.113a	2.35	6.42a	0.051b	0.036b	3.20a	10.4	24.7
Zr-95	0.397b	0.314a	NR							0.150b	NR
Rh-103	10.9a	17.6a	8.79a	0.052c	1.29a	NR	0.056d	0.291c	1.66a	4.81a	12.1
Cs-137	32.1	48.1	21.9	0.158	3.70	7.72	0.105a		6.17	18.7	39.4
Ce-144	75.8	103a	53.7	0.281a	8.18	16.9	0.192b	0.150a	11.6	35.7	68.9a
W-181	324c	244	119		22.8a	53.7			50.9b	122b	335
W-185	313a	335a	NR		NR	NR			91.3a	200a	NR
Pb-210	0.503	0.661		0.111	0.446a			0.019a	0.316	0.421	

TABLE VI (Cont'd)

ATMOSPHERIC RADIOACTIVITY DATA, NOVEMBER 1960

Date 1960	Nov 1	Nov 10	Nov 15	Nov 1	Nov 10	Nov 15	Nov 18	Nov 21		Nov 2	Nov 8	Nov 15
Latitude	71°N	72°N	71°N	36°N	33°N	34°N	8°N	9°N		63°S	6 °S	62°S
Longitude	164°W	164°W	164°W	105°W	105°W	105°W	158°W	158°W		167°E	16 °E	167°E
Altitude (Ft.)	50,000	50,000	50,000	50,000	50,000	50,000	50,000	50,000		50,000	50,000	50,00
Aircraft Type	U-2	U-2	U-2	U-2	U-2	B-57	U-2	U-2		U-2	U-2	B-57
Exposure (Minutes)	296	368	300	159	313	129	305	312		360	35	111
Volume (1000 SCF)	224	282	229	133	263	149	220	224		237	287	121
Tropopause (Mt., Ft.)	34,100	32,300	29,000	54,000	39,000	36,700	53,000	54,500		36,000	29,500	33,000
Isotope												
Be-7	379	698a	758	65.8a	201a	559c	62.6a	20.7b		302	333	636
Sr-90	35.5	111	99.5	1.66a	9.90	25.5	0.740a	0.205b		26 8	26.	41.5
Zr-95	0.593b	1.82a	1.50			NR				0.381a	0.326a	NR
Rh-102	20.8a	116	39.8a	1.59a	5.15	15.7a	0.376b	0.109d		14.0a	9.59a	20.8
Ce-137	64.5	190	155	2.82	16.9	36.6a	1.19	0.349a		47.7	45.9	59.3a
Ce-144	143	529	410a	5.41	35.6	80.4	2.79	0.886		89.5	87.3	120
W-181	361a	351a	454b		111b	133a				420a	375b	518
W-185	368a	584	NR		176	NR				NR	525a	NR
Pb-210	0.520	0.225a		0.309	0.512a		0.286a	0.152a		0.293	0.326	

Date 1960	Nov 1	Nov 10	Nov 15	Nov 1	Nov 10	Nov 16	Nov 19	Nov 21	Nov 24	Nov 2	Nov 8	Nov 15
Latitude	71°N	71°N	72°N	36°N	36°N	33°N	9°N	8°N	9°N	63°S	61°S	61°S
Longitude	164°W	164°W	164°W	105°W	105°W	105°W	158°W	158°W	158°W	167°E	167°E	167°E
Altitude (Ft.)	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000	60,000
Aircraft Type	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2
Exposure (Minutes)	238	361	366	345	342	287	316	351	347	360	184	319
Volume (1000 SCF)	103	156	159	150	148	124	127	142	140	148	78.5	147
Tropopause (Mt., Ft.)	34,100	32,300	29,000	54,000	39,000	64,000	53,000	54,500	54,600	36,000	29,500	33,000
Isotope												
Be-7	597a	698b	1116	553a	797	557a	324a	385	251	726	711a	939
Sr-90	129	111	145	85.1	130	84.4	37.2		26.8a	138	137a	150a
Zr-95	2.19b	2.19a	1.55	1.52	2.28a	1.02	0.920a			2.71a	2.96a	NR
Rh-102	112	203a	134	72.2	150	316a	24.0a	19.8b	9.94	178	159	46.8c
Ce-137	221	190	212	135	211	127	67.5	38.8	44.7	246	217	226a
Ce-144	586	602	626	358	624	324a	157	78.8	96.0a	583	536	416
W-181	512b	88.3c	366	414c	432b	563	378d	305c	NR	741a	857b	456
W-185	859a	234d	NR	649a	637b	NR	475b	576	NR	1079a	1239a	NR
Pb-210	0.301a	0.090a		0.356	0.271a		0.566	0.617a		0.233a	0.312b	

Date 1960	Nov 1	Nov 10	Nov 15	Nov 1	Nov 10	Nov 15	Nov 18	Nov 22	Nov 24	Nov 2	Nov 8	Nov 15
Latitude	71°N	72°N	71°N	32°N	33°N	32°N	9°N	8°N	9°N	60°S	60°S	60°S
Longitude	164°W	164°W	164°W	105°W	105°W	105°W	158°W	158°W	158°W	167°E	167°E	167°E
Altitude (Ft.)	63,900-65,600	62,000-64,000	61,900-64,600	65,000-67,300	65,000-67,400	66,000	65,900-68,600	64,500-67,900	65,700-68,900	60,000-64,000	62,000-64,000	62,000-67,000
Aircraft Type	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2	U-2
Exposure (Minutes)	352	387	367	373	436	459	308	310	320	421	326	404
Volume (1000 SCF)	107	133	123	101	119	126	77.0	84.2	80.0	156	111	124
Tropopause (Mt., Ft.)	34,100	32,300	29,000	54,000	39,000	64,000	53,000	51,500	54,600	36,000	29,500	33,000
Isotope												
Be-7	655a	865	1462b	792	591	563	267	220	296b	553a	727	797
Sr-90	148	147	168	142	136	139a	71.8	62.3	83.1	137	157	173a
Zr-95	1.86b	2.04a	1.50	2.94a	1.89a	NR	NR	1.37b	NR	2.50	2.36	NR
Rh-102	186	262b	261b	178	133	737a	47.3	55.3b	NR	151	206a	240
Ce-137	258	239	245	239	227	231a	138	112	145	235	276	285a
Ce-144	748	822	744	726	614	578	265	217	261a	569	652	589
W-181	193c	93.3b	254a	428b	383a	436	974	928a	1131	543b	NR	402a
W-185	403b	NR	NR	779a	635a	NR	1266	NR	NR	846a	753a	NR
Pb-210	0.214a	0.111a		0.243a	0.271a		0.636a	0.607a		0.166a	0.236a	

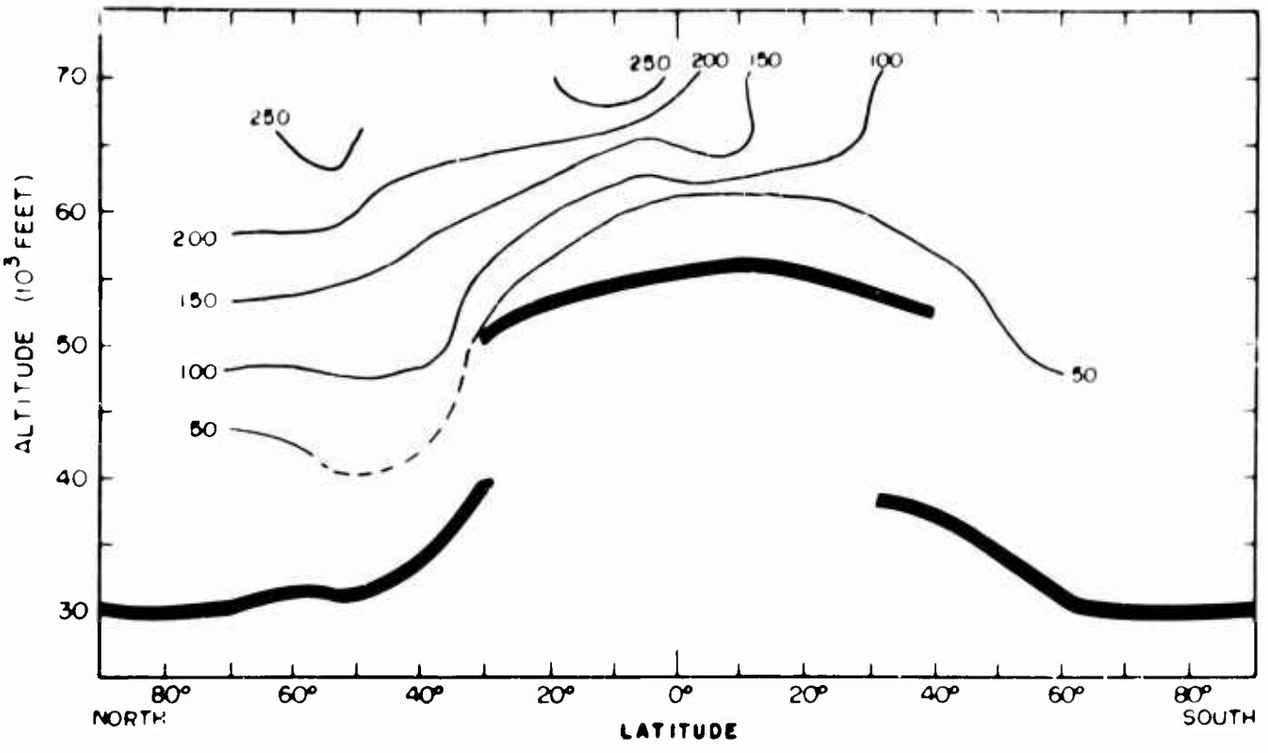


FIGURE 15a STRATOSPHERIC DISTRIBUTION OF STRONTIUM - 90 (dpm / 10^3 SCF) DURING MAY - JUNE 1959

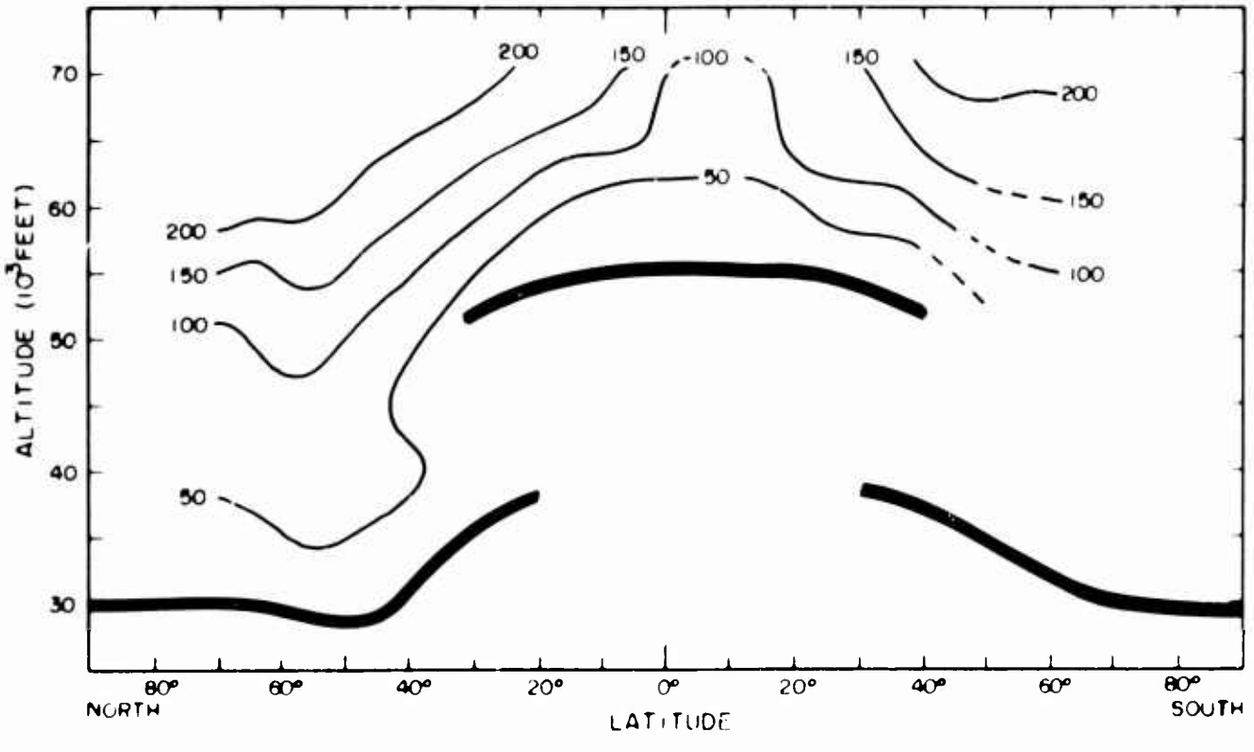


FIGURE 15b STRATOSPHERIC DISTRIBUTION OF STRONTIUM - 90 (dpm / 10^3 SCF) DURING MAY - JUNE 1960

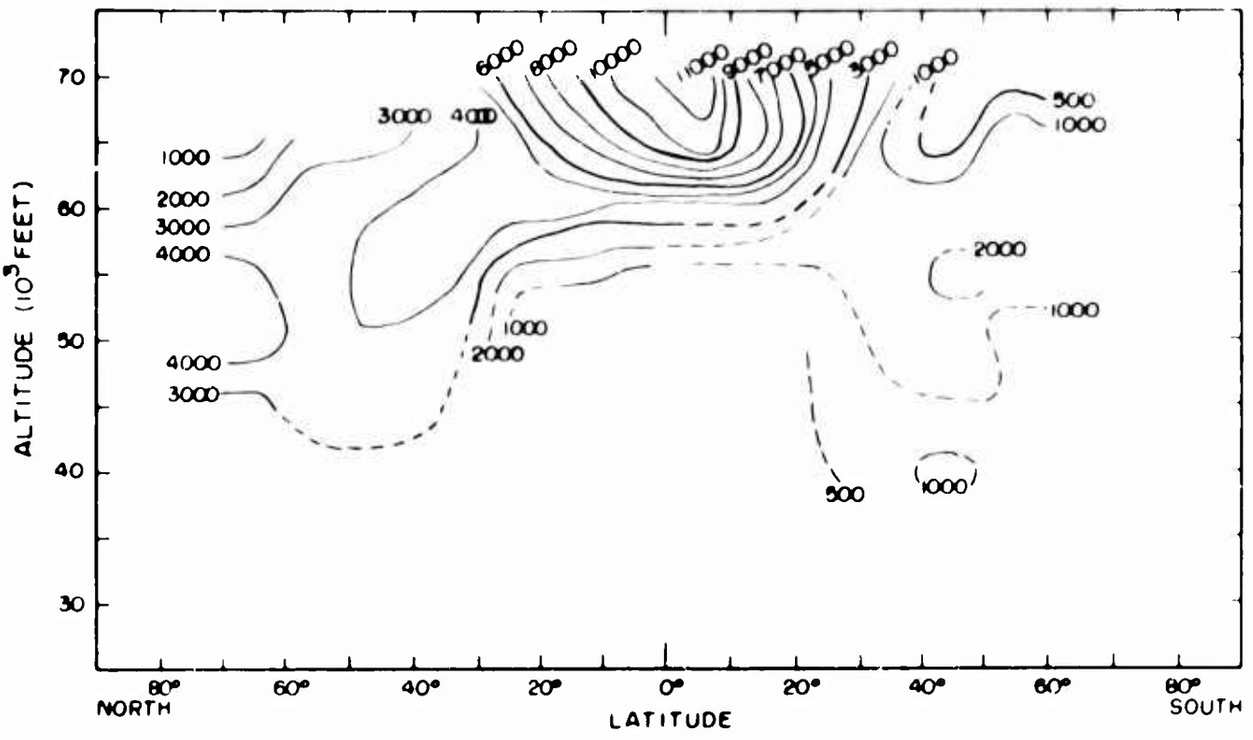


FIGURE 16a STRATOSPHERIC DISTRIBUTION OF TUNGSTEN-185 (dpm/10^3 SCF corrected to 15 Aug 1958) DURING MAY-JUNE 1959

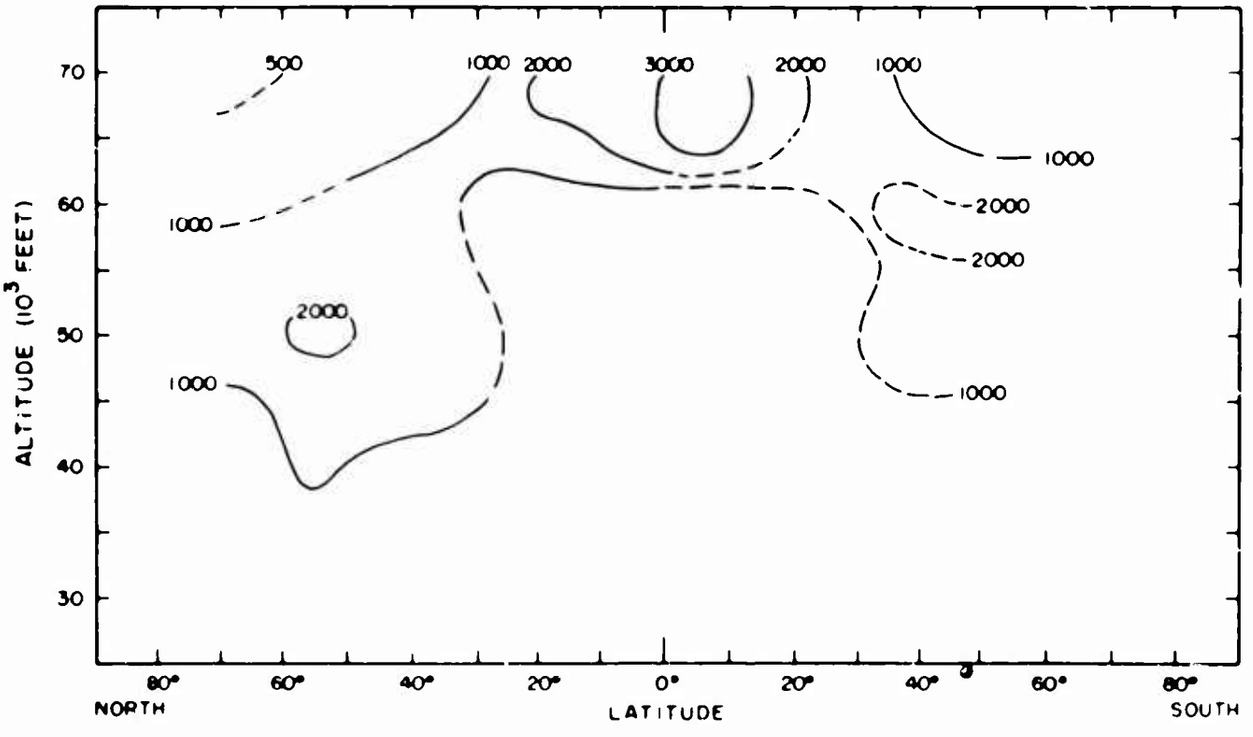


FIGURE 16b STRATOSPHERIC DISTRIBUTION OF TUNGSTEN-185 (dpm/10^3 SCF corrected to 15 Aug 1958) DURING MAY-JUNE 1960

in which tungsten is distributed. First the equatorial maximum at 70,000 feet has remained stationary although its value has diminished. The equatorial values of tungsten were reduced by about 70% in one year while the strontium-90 values in the equatorial region were reduced only by about 50%. This could be accounted for by the originally steeper tungsten gradient and by the fact that some strontium-90 could have mixed from the new polar maxima toward the equator. In the polar regions the tungsten maxima occur well within the sampling region and the tungsten values diminish at the higher altitudes while the strontium-90 values increase at the higher altitudes.

These distributions can be best explained by the mechanism of turbulent diffusion along surfaces sloping downward from the equatorial region. The drop off of tungsten at the high altitudes and at high latitudes indicates that most of the stratospheric tungsten was originally introduced into the lower tropical stratosphere (below 80,000 feet). The tungsten distribution provides a very strong argument against the Brewer-Dobson circulation model.[15] Further discussion of this material is contained in Chapters V, VIII, and IX.

The cerium data indicates that the youngest debris can be found at the highest altitudes and highest latitudes again pointing to Teak and Orange debris. Concentrations of Rh-102, a tracer element placed in the Orange shot, are highest at the highest altitudes and highest latitudes. These distributions indicate that material injected into the atmosphere above 200,000 feet follows a downward route to the lower stratosphere preferentially in the polar regions. Further discussions on these points will be found in Chapters VI, VIII, and IX. The cesium-137 data, in general, follows

the strontium-90 data in concentrations of about 1.8 times the strontium-90 values. Except for an evaluation of the hazard from cesium-137 in Chapter XI, little further discussion of this isotope will be presented in this report.

The Be-7 data is symmetrical between hemispheres and shows a maximum at high latitudes and high altitudes. The gradients are not nearly as steep as those of rhodium-102, however, and reflect the cosmic ray origin of Be-7. The P-32 data is helpful in determining the age of the Be-7. In general the distributions of these two nuclides are about as expected (see Chapter VII). The Pb-210 distribution in the stratosphere is much like the tungsten data and suggests an equatorial stratospheric origin for most of this material. Although Pb-210 is found in the troposphere as a decay product of radon, it is unlikely that the stratospheric distribution observed comes from this source alone, but rather that some of it was produced during the weapons testing programs of the past decade. An analysis of some of the older filter papers in the HASP library, which is now being undertaken, should be able to resolve this point. Preliminary results indicate that if the equatorial Pb-210 is of weapons test origin, it predates the HARDTACK series. A more detailed discussion of the Pb-210 data is given in Chapter VII.

Additional comments on the data in Table VI have been prepared in the U. S. Weather Bureau.[38]

Chapter V

STRONTIUM-90 AND TUNGSTEN-185 IN THE STRATOSPHERE

Introduction

For the past few years strontium-90 and tungsten-185 concentrations in the lower stratosphere have been measured over a wide range of latitudes by the HASP and Ashcan programs. These measurements have been helpful in determining the total stratospheric burden of these elements and other relatively long lived fission products. In addition, considerable information has been gained on the mixing and transfer of these fission products within the stratosphere.

While the HASP network has been successful in many areas, it has suffered from several deficiencies. The greatest of these is the inability of the U-2 to collect samples above 70,000 feet. Estimates of concentrations above this altitude have been made using data from the AEC Ashcan Program.[26,27] This program, too, has an altitude deficiency in that collections are limited to below 100,000 feet. To date, no measurements of radioactive weapons debris in the mesosphere (160,000-260,000 ft.) or above have been reported. In addition to the altitude limitations mentioned above, both the HASP and Ashcan networks suffer from lack of latitudinal coverage. Few measurements have been made in the highest latitude polar stratosphere, and in the case of the Ashcan samples, collections have been restricted to 1 or 5 geographical points. In addition, the time coverage has been somewhat spotty. HASP samples north of 10^{o} N during the winter of 1958-1959 were few as were samples south of the equator after August 1959. Ashcan sampling in the Southern Hemisphere ceased on 7 February 1959.

One final complicating factor should be mentioned and that is the repeated and continuing influx of radioactive material into the sampling volume from adjacent

regions. The primary source of this influx has been, of course, the stratospheric injections from atomic tests which have been conducted since the sampling program began. Accurate appraisals of the quantity of material injected into the stratosphere is difficult and estimates of vertical distribution remain somewhat speculative. Another perturbing influx of debris has been that which has mixed downward from the Teak and Orange events. While these events have somewhat obscured the calculation of the atmospheric burden of various radioisotopes, they have provided the concentration gradients necessary to determine the stratospheric mixing processes. Considerable progress has been made during the quiet period since late 1958.

Distribution of Strontium-90

Attempts have been made to determine the average strontium-90 burden in the stratosphere during several time intervals. The method has been described in the previous report (DASA-532). Various small regions in the atmosphere have been selected and assigned an average strontium-90 concentration based on a number of sample collections made in the region during the time period under consideration. Extrapolation poleward has been done on the assumption that high latitude concentrations have been linear extensions of the lower latitude concentrations. Extrapolation upward from the HASP sampling region has been done using the shape of the Ashcan profile. A 0.03 micron particle efficiency in the Ashcan sampler has been assumed in order to cause the HASP and Ashcan 65,000 feet measurements to overlap. The average values can be arranged to show the meridional distribution by plotting on a vertical cross-sectional representation of the atmosphere and then drawing iso-intensity contour lines (isopleths) to encompass those points in space where the strontium-90 concentrations are the same. Figures 17 through 20 show,

respectively, the strontium-90 concentrations for 1958, early 1959, late 1959 and early 1960.* The first three charts were drawn using a vertical scale showing a linear decrease in ambient pressure (inverse log of altitude) and a horizontal scale proportional to the sine of the latitude. This depiction facilitates measuring the inventory since equal areas on the chart correspond closely to equal volumes of air. A planimeter may then be used to measure the volume of the atmosphere encompassed by each isopleth. The tacit assumption is made that all meridians have the same profile shown in the charts. All isopleths are expressed in decays per minute per 1000 standard cubic feet of air sampled. Reduction to standard sea level pressure is made to remove the altitude and temperature dependence of air density. Concentrations above the level of the Ashcan sampler have been assumed to drop off to zero linearly and no attempt has been made to indicate possible concentrations in the mesophere (see Chapter VIII). Figure 20 differs from Figures 17-19 in that the scales are linear in altitude and latitude and do not extend above the HASP sampling level. This altitude limitation is placed on the chart because no extensive data above 70,000 feet are available for the period in question. Consequently any isopleths above this level would be most conjectural.

The most prominent features of the 1958 strontium-90 distribution (Figure 17) are the maxima in the equatorial regions and northern polar regions as a result of the US-UK, HARDTACK-GRAPPLE test series of 1958 and the autumn USSR test series of 1958. By mid-1959 (Figure 18) these maxima gave the appearance of having merged and the concentration gradients had become less steep. Little change in the Northern Hemisphere was noted during the last half of 1959 (Figure 19). This was reflected

*All isopleths shown are based on old flow data, but the hemispheric burdens of strontium-90 indicated in Table VII have been recalculated using the net flow rates reported in Chapter II.

in the lowered surface fallout rates noted in ground sampling networks at this time (see Chapter X). There is no strong evidence of substantial amounts of debris from the USSR test series of 1958 remaining in the stratosphere by late 1959. Presumably the major fraction had been removed within one year. This is confirmed by age studies (Chapter VI) and surface measurements (Chapter X).

By the spring of 1960, the strontium-90 picture had changed from one of high concentrations at the equator and low concentrations at high altitudes in the polar regions to one of just the reverse. The increasing gradient poleward and upward is believed to be due to the influx of debris from the Teak and Orange shots of HARDTACK moving down into the sampling region at high latitudes, and not due to "Brewer-Dobson" circulation. This belief is strengthened by the tungsten data reported below.

The low concentrations at high altitude in the polar regions in 1958 indicate either that the CASTLE and REDWING debris has not mixed into the polar regions or that it has already mixed into and subsequently been removed from this area. The rates of equatorial-polar transfer to be described below are sufficiently rapid that it seems unlikely that this debris has been held up in the equatorial region. Indeed, there has been little evidence of the presence of any large cohesive concentrations of CASTLE or REDWING debris within the HASP network since the sampling program began in late 1957.

Strontium-90 Burden

An attempt has been made to calculate the burden of strontium-90 in the atmosphere above the tropopause. (The burden in the troposphere within a few months after nuclear detonations is quite low compared to that of the stratosphere due to the relatively short tropospheric residence time.) This may be done by starting with

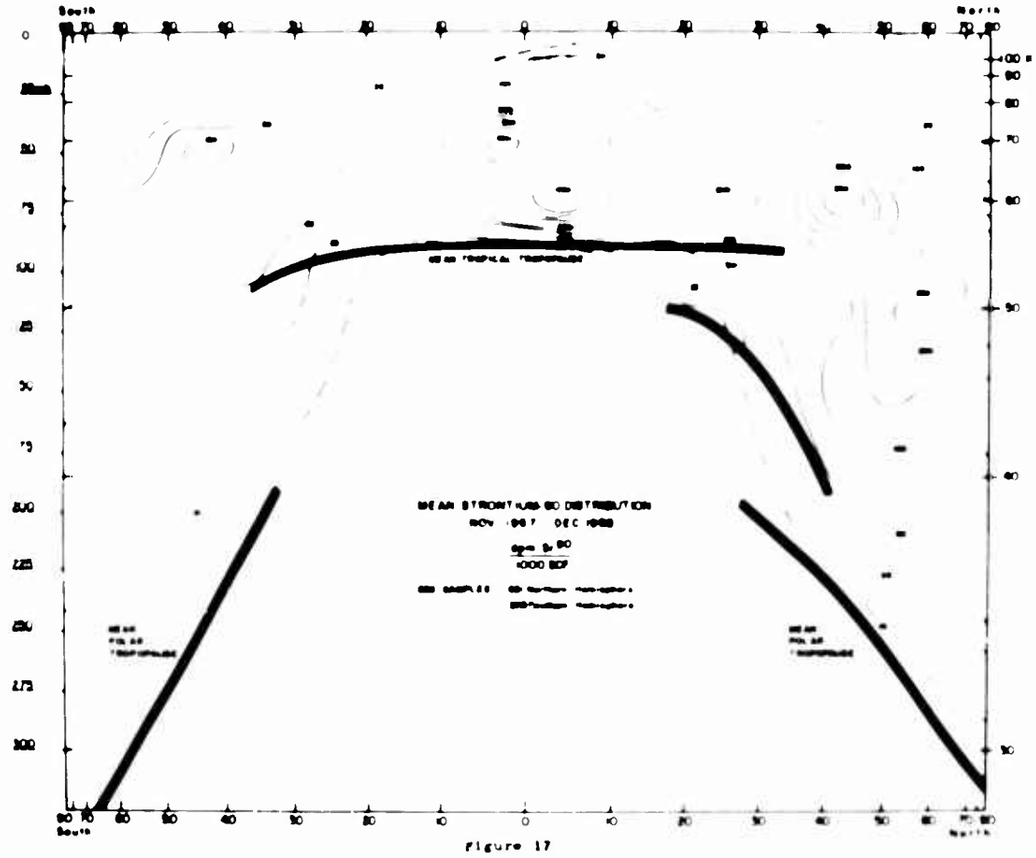
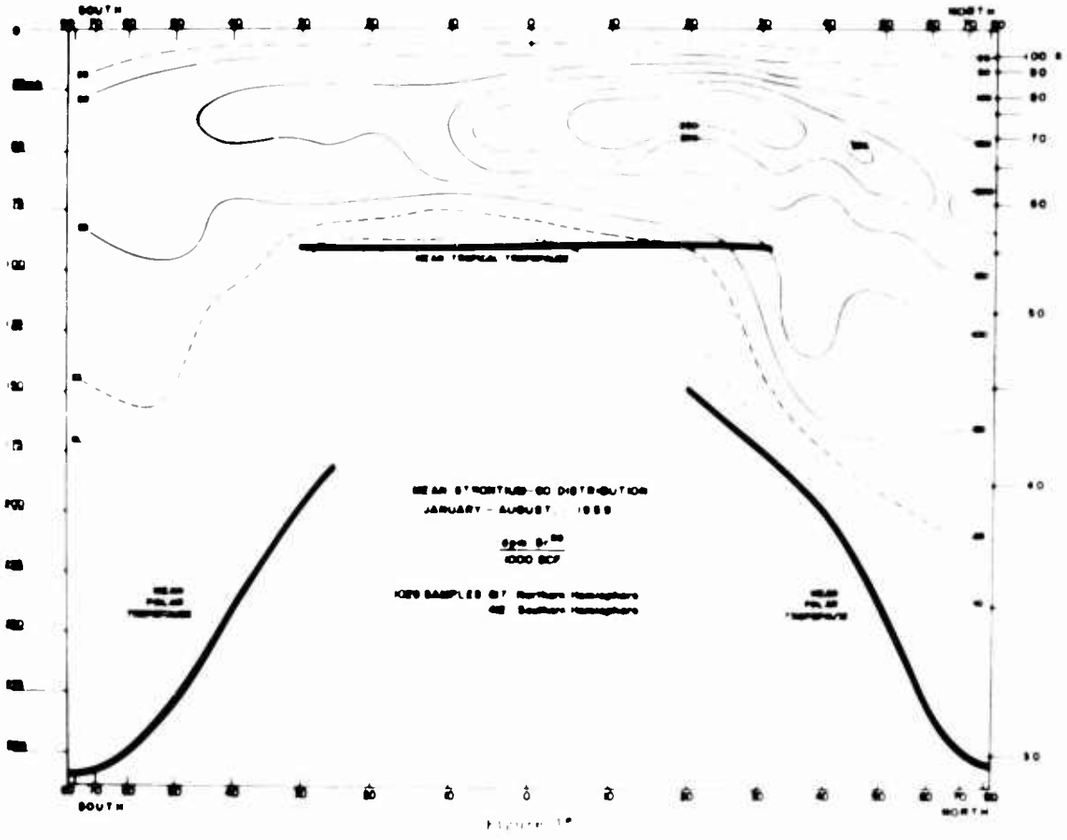


Figure 17



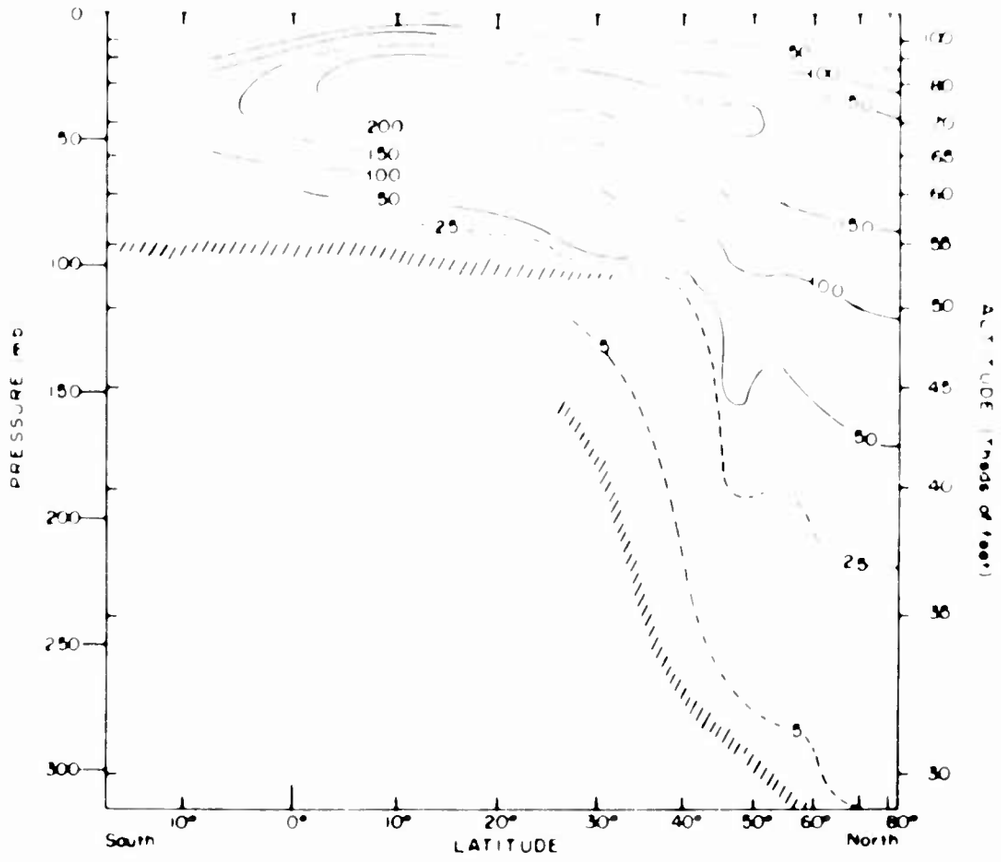


FIGURE 19 MEAN DISTRIBUTION OF STRONTIUM-90 IN THE STRATOSPHERE, SEPTEMBER-DECEMBER, 1959 (779 Samples)

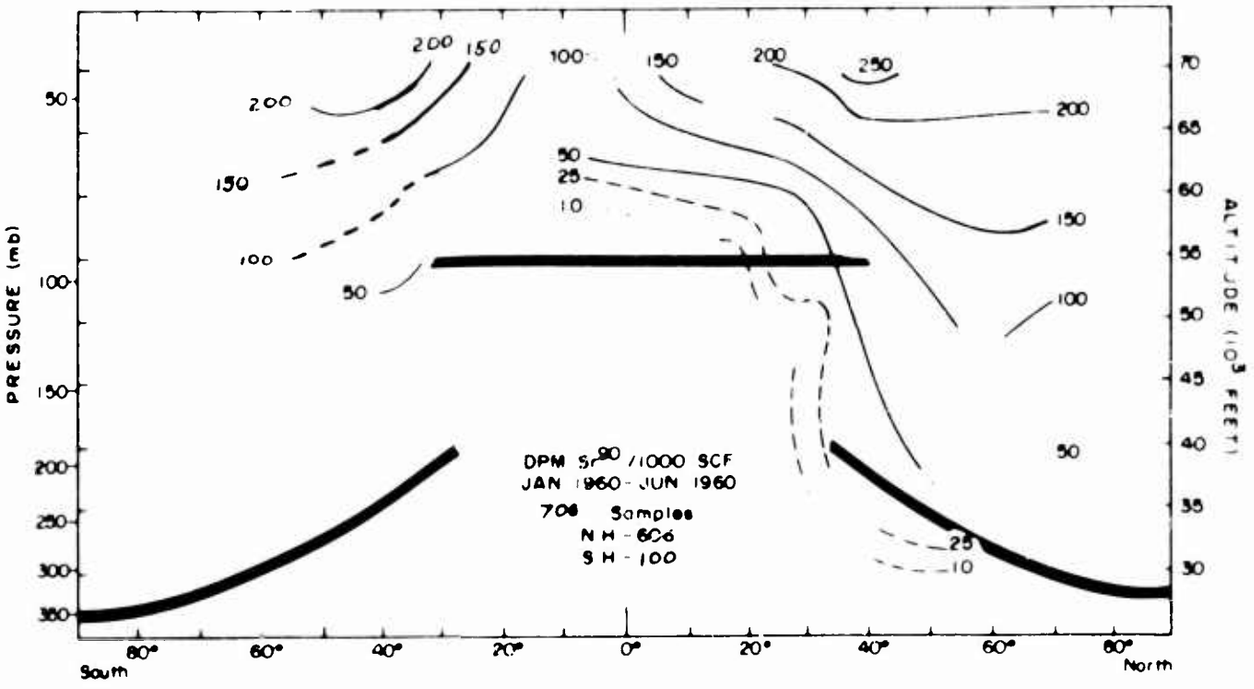


Figure 20 MEAN DISTRIBUTION OF STRONTIUM-90, JANUARY 1960 - JUNE 1960

the measurements in the HASP sampling area and integrating these values in the manner described in the preceding section. This will give measurements up to about 50 mb (70,000 ft.) with about 15% accuracy. The Ashcan measurements can be used to extrapolate upward to about 10 mb (100,000 ft.) with perhaps 30% accuracy. Extrapolations above 10 mb must remain somewhat conjectural as no measurements in this region have been reported. It has been estimated[28] that as much as 0.1 megacuries of strontium-90 have been injected into the mesosphere from nuclear tests (see Chapter VIII). Extrapolations across gaps in time and into unsampled areas can be made using assumed initial burdens and apparent residence times calculated or deduced from other evidence. Calculations of this nature have been made and the results are shown in Table VII below. It should be pointed out that there is a relatively large element of subjectivity in establishing a single number to represent the average value of the strontium-90 concentration in a large region over a period of time as long as one year. This is especially true for the period 1958-1959 when there were large changes in certain regions due to the extensive injections from weapons testing activity and the dynamic processes of mixing between separate regions. The inherent uncertainties are compounded by the fact that the debris was not distributed zonally in a symmetric fashion during times of active injection and, in addition, large quantities of Soviet debris escaped adequate measurement due to its altitude and latitude of stabilization as well as its relatively short residence time. Undoubtedly there were times during the periods under consideration when the inventories were substantially greater or less than the values shown in Table VII. (See Fig. 52 Chapter IX). A material balance of strontium-90 based upon these estimates and soil and rainfall measurements is reached in Chapter X.

TABLE VII

Crude Strontium-90 Inventories (in Megacuries)

Northern Hemisphere

Height	Nov 57-Dec 58	Jan 59-Aug 59	Sep 59-Dec 59	Jan 60-May 60
0 mb - 10 mb	.20 ± .01	.20 ± .01	.17 ± .01	.13 ± .01
10 mb - 50 mb	.15 ± .05	.27 ± .08	.30 ± .08	.25 ± .10
50 mb - Trop	.61 ± .09	.38 ± .06	.33 ± .05	.37 ± .06
Total	.96 ± .20	.85 ± .16	.80 ± .15	.75 ± .15

Southern Hemisphere

0 mb - 10 mb	.20 ± .01	.19 ± .01	.16 ± .01	.12 ± .01
10 mb - 50 mb	.11 ± .03	.15 ± .05	.17 ± .06	.13 ± .05
50 mb - Trop	.22 ± .03	.13 ± .02	.16 ± .08	.27 ± .08
Total	.53 ± .13	.47 ± .10	.49 ± .13	.52 ± .13

World Total

.49 ± .33	1.32 ± .26	1.29 ± .28	1.27 ± .28

The heavy line in Table VII separates the "measured" values from the "estimated" values, the values below the line being those calculated from HASP and Ashcan data. The table shows that the burdens in the Northern and Southern Hemispheres have tended to equalize. This has been brought about by the rapid fallout of the Soviet debris in the Northern Hemisphere. In addition there has probably been some diffusion across the equatorial region caused by the horizontal gradient in this area (See Figure 19). The rise in the total burden in the Southern Hemisphere during the last half of 1959 is probably more apparent than real. The only measurements made well into the Southern Hemisphere were performed during Phase V below 50 mb and were of short

duration. The errors of measurement are such that the total burden could have actually gone down. Another feature noticed is that the net residence time of the debris in the stratosphere appears to have increased. This is due to the increase in relative amounts of material at the higher altitudes where residence times are expected to be longer.[2]

Stratospheric Mixing of Strontium-90

Considerable information can be gained on stratospheric mixing processes by observing the monthly strontium-90 concentrations at several latitudes and altitudes. Figure 21a illustrates the time variation of strontium-90 concentrations for three selected latitudes.

Equatorial Region

Between 10^{o} and 15^{o} North, the latitude of the Pacific tests, several features can be noted. First the vertical gradient is quite pronounced and remains more or less constant with time indicating that vertical mixing is slow. Second, the concentration at each altitude is diminishing with time after September 1958 showing a departure of debris from this area. Most of this debris probably moves poleward as will be shown below. Third, the rate of departure of the debris is not uniform. Removal is seen to be fast during the winter and spring, and slow or even halted during the summer and fall. This seasonal dependence upon rate of movement from the equatorial regions contributes to the spring maximum noted in surface fallout by increasing the concentration of material in the polar stratosphere during the winter. The increased rate of poleward movement is probably associated with the intensification and equatorward movement of the mid-latitude jet stream. During May 1960 there has been an apparent rise in concentration. This may be due to some material from Teak and Orange reaching this latitude although, by itself, this evidence is not conclusive.

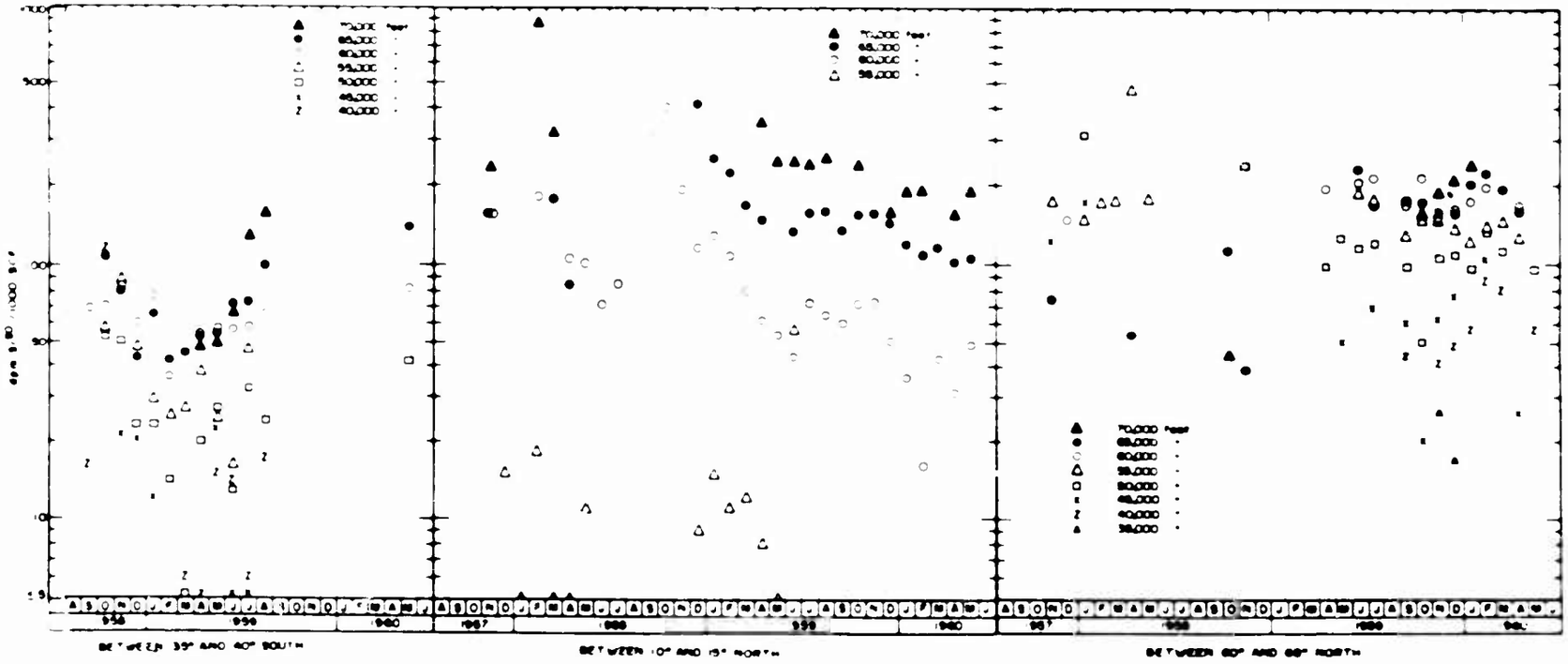


Figure 21a

North Polar Region

Between 60^{O} and 65^{O} North a considerably different picture has prevailed. Until after November 1958 a negative vertical gradient existed at this latitude. Some evidence exists in the HASP samples which collected debris from the Soviet tests that these clouds did not rise above 65,000 feet, consequently the concentration at 60,000 feet was higher than at 70,000 feet. There was also evidence that the Soviet clouds sampled over Canada were found considerably further South than the Arctic test site indicating an asymmetry in the circumpolar flow at that particular time. During the spring of 1959, the Sr-90 concentrations rose, due to an influx of HARDTACK-GRAPPLE debris, and then fell off slightly during the summer.

Starting in October 1959 the concentrations in the northern polar latitudes started to increase at 70,000 feet. At this same time the concentrations at lower altitudes were still decreasing. By December the concentrations at 65,000 and 60,000 feet had started to increase. Concentrations at 55,000 and 50,000 feet did not start to increase until January 1960. This behavior is strongly indicative of an influx of material from above into the polar region. This could have resulted from two sources. Either the material mixed northward from the equatorial region or it mixed downward from Teak and Orange in the arctic region. Probably it was a combination of both. The polar increase is consistent with the seasonal equatorial depletion described above. Cerium dating evidence (Chapter VI) indicates that a large fraction of this increase may have come from Teak and Orange. If the rise in concentration were due mostly to influx of HARDTACK GRAPPLE debris then the 10^{O} to 15^{O} North concentrations should show a similar but earlier increase. An increase is observed in this region but it is not as pronounced as that at the 60^{O} to 65^{O} North region.

One feature of note which may be seen at this high latitude is that the seasonal influx appears to reach a maximum in the lower polar stratosphere during the winter. This can account for the spring maximum noted in surface fallout about April. An additional feature which is quite noticeable in the polar stratosphere is that the vertical concentration gradient is not nearly as steep as that observed in the equatorial regions. This indicates that vertical mixing in the polar stratosphere proceeds at a much higher rate than in the equatorial regions, especially during the winter.

Ashcan Data

The Ashcan data for San Angelo, Texas, and Sioux City, Iowa, for 1959 show certain similarities to the HASP data. Figure 21b shows the averaged Ashcan data for several years as depicted by Murayama and Machta.[39] There appeared to be a gradual diminution at all altitudes suggesting a half residence time of about one year. The maximum concentration over San Angelo occurred at about 80,000 feet until the winter of 59-60. At this time the maximum shifted downward and values at 65 to 70,000 feet showed a marked increase, more than noted in the HASP samples in the same area. The data suggests an increased influx from the tropical region along a downward sloping mixing layer especially during the winter of 1958-59. This kind of mixing is especially prominent in the tungsten data discussed below. The Sioux City data shows a less pronounced vertical gradient than that at San Angelo. This is consistent with the HASP data discussed above which suggests stronger vertical mixing in the polar stratosphere. The Ashcan data, in general, shows a much greater monthly variability than does the HASP data. This is probably due primarily to the small sample taken in a relatively restricted area. A whole Ashcan sample typically measures 1000 scf while the typical HASP sample contains 10,000 to 20,000 scf. In

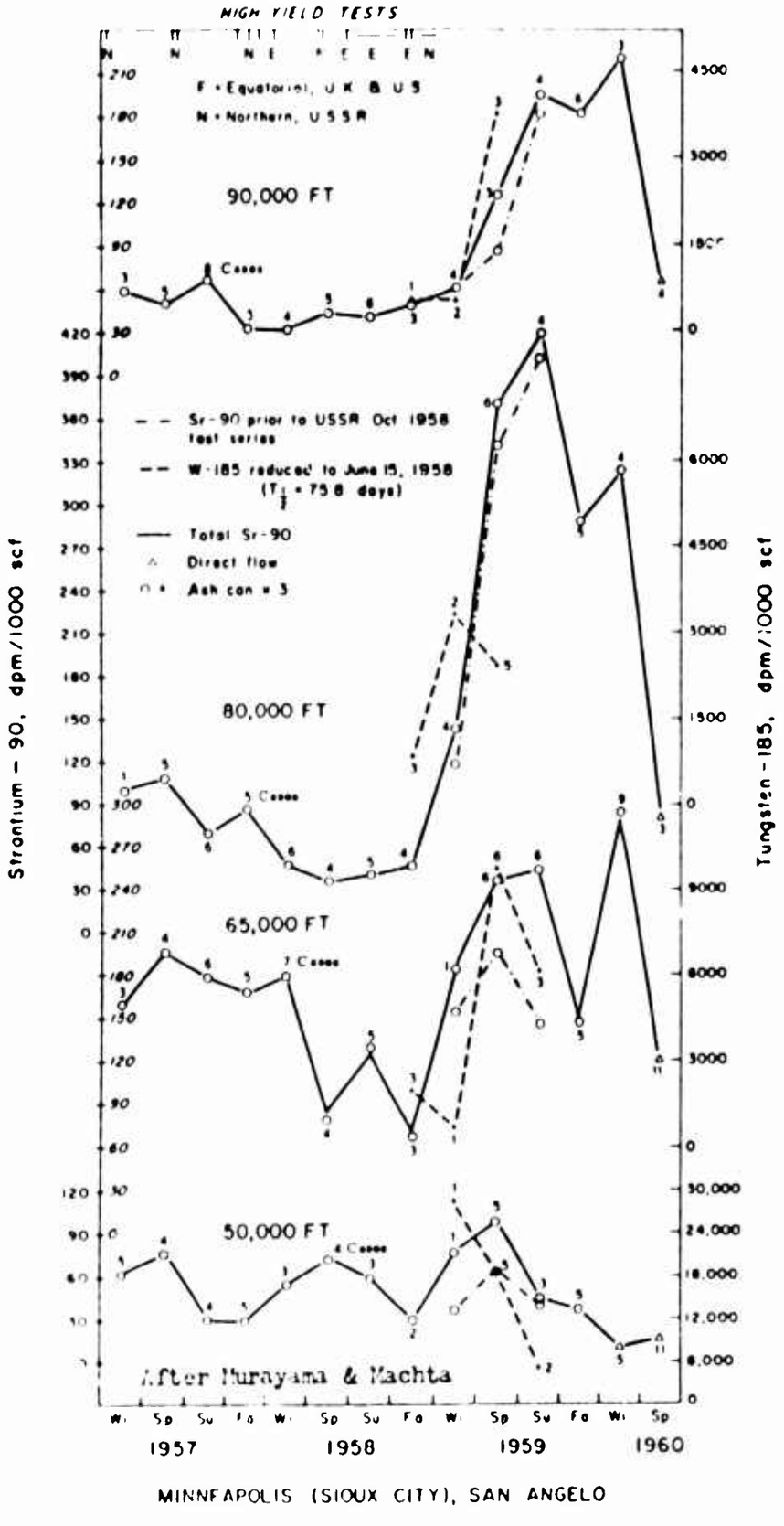


Figure 21b Averaged Ashcan Data

addition, a number of HASP samples are averaged for each location in a month while the Ashcan generally collects but one sample per altitude per month at each station. Another problem with the Ashcan samples is the relatively low efficiency of collection which is not only altitude dependent but also particle size dependent. Only general trends can be noted with any reliability in the Ashcan samples.

South Polar Region

In some respects the concentrations in the southern polar stratosphere (35° - 40° S) are similar to those of the northern polar stratosphere. During the spring, summer and early autumn of 1958-59 (October 1958 - May 1959) concentrations generally were at a maximum near 60,000 feet showing that HARDTACK-GRAPPLE debris preferentially mixed downward into this region along sloping surfaces much like those followed by the HARDTACK tungsten debris (see below). During the winter of 1959 (May through August) concentrations increased markedly especially at the higher altitudes. Presumably this was caused by an increasing influx from the higher equatorial regions at that time. Cerium dating of this material indicates that it is a little older than that found in the lower tropical stratosphere; consequently, it is felt that only a small fraction of the increase could have come from Teak and Orange. Some rhodium-102 was produced in the low altitude equatorial shots of HARDTACK and was found to be distributed in both hemispheres in the same manner as was tungsten.[2] However, somewhat larger quantities of rhodium-102 were found in the southernmost and highest papers collected in August 1959 (late winter). This is considered to be the first piece of evidence that material from Teak and Orange had reached the lower stratosphere.[29,30] By the next winter (June 1960) concentrations had increased about 50% at all altitudes above 50,000 feet. In addition, the age of the debris had become younger indicating a further incursion of Teak and Orange debris. The

vertical gradient seemed to be somewhat steeper than that of the northern polar stratosphere. This may be partially due to the fact that the southern latitude chosen for comparison is closer to the mid-latitude tropopause break where faster influx of clean air may occur at the lower altitudes.

Distribution of Tungsten-185

During the course of the HARDTACK testing in 1958 several weapons were fired which produced large quantities of tungsten-185 (as well as other tungsten isotopes). In addition, a few of these weapons had sufficient total yield to inject a large fraction of the activity into the stratosphere. The production of tungsten-185 has been variously reported.[2,31,32] Lockhart[31] suggests that production in thermonuclear weapons can run as high as four W-185 atoms produced per 10 fissions. This would yield a W-185/Sr-90 activity ratio of about 1800. The amount measured in the stratosphere would depend upon the number of weapons burst that produced tungsten compared to the total, the yield spectrum for each, and the time intervals between bursts. In addition, the selection of a reference date to compare tungsten and strontium values would affect the apparent ratio. The values noted in debris from a tropospheric source could be expected to vary substantially from that coming from a stratospheric source. Lockhart[31,33] has used a W-185 Sr-90 ratio of 378 as of 15 August 1958 to categorize HARDTACK fallout. Hardy[34] has used a value of 180 while Martell[32] has used a value of 216 for this ratio as of 15 August 1958. The previous report (DASA-532) estimated an effective stratospheric burden as of 15 August 1958 to be about 53 megacuries. This is somewhat lower than the amount reported by Martell but seems to be consistent with the HASP inventory and the total inventory of 50 megacuries (corrected to 15 August 1958) through 1959 in rainfall at the surface calculated by Hardy.[34] Martell's value of 216 for the W-185 Sr-90 ratio, however,

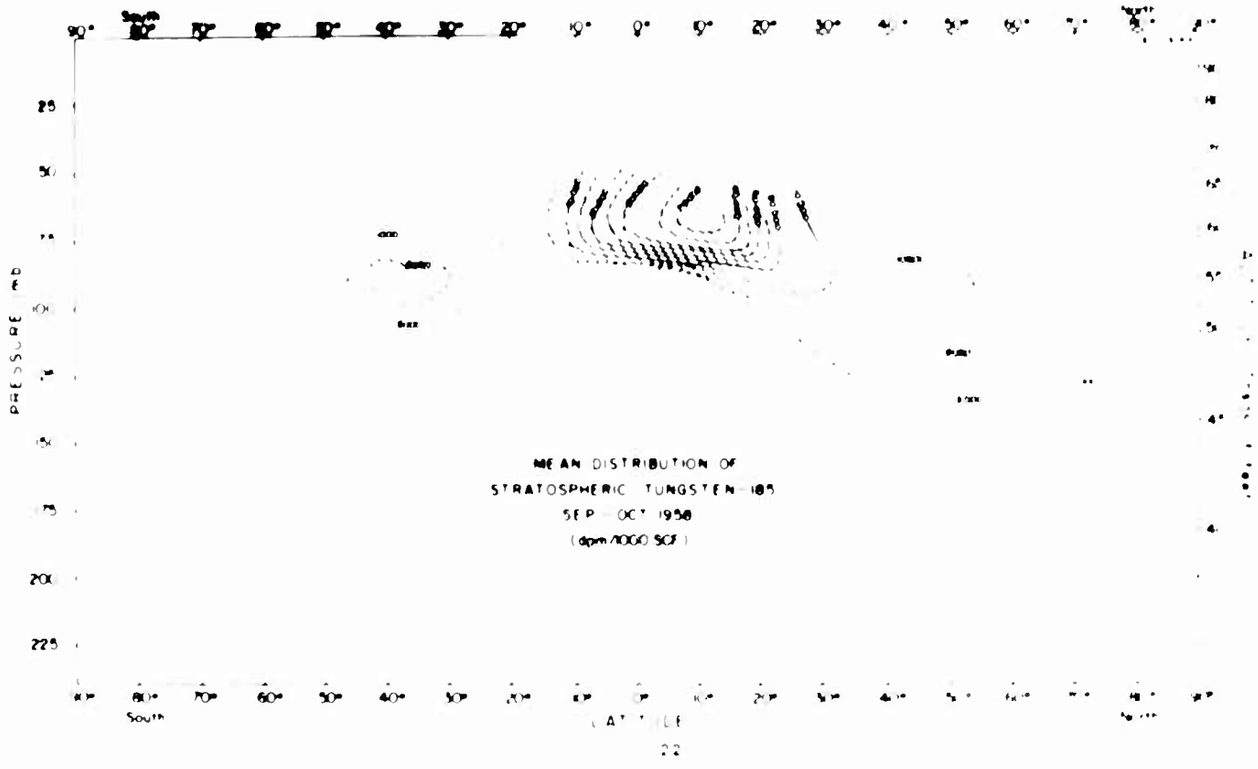
is in agreement with the amount of strontium-90 and tungsten injected during HARDTACK (exclusive of Teak and Orange) when calculated by the method described in the previous report.[2]

The material from the UK GRAPPLE series, which showed no tungsten, has diluted the HARDTACK debris as far as the total effective W-185/Sr-90 ratio for the equatorial region is concerned. A value of 85 for this ratio as of 15 August 1958 may fairly well account for the total stratospheric tungsten-185/strontium-90 ratio.* This value is in fair agreement with the Porto Alegre, Brazil average of 9. ± 17 during July 1958-March 1959 which would be expected to show a HARDTACK-GRAPPLE mixture as suggested by Martell.[32] It should be pointed out that the average height of injection of tungsten was slightly lower than the average height of injection of Sr-90 from HARDTACK-GRAPPLE. This can be seen by comparing Figure 17 with the tungsten figures that follow. As a consequence the fallout of HARDTACK tungsten may not be a perfect way to categorize either the total HARDTACK injection or the combined HARDTACK-GRAPPLE injection. However, since tungsten is a unique tracer not obscured by a previous background it serves as an ideal vehicle to study transfer of lower stratospheric air from the equatorial region to other regions.

Figures 22 through 31 show bimonthly plots of tungsten-185 concentrations from September 1958 through April 1960. All except Figure 31 are plotted with a linear latitudinal scale and a linear barometric pressure scale. Figure 31 is plotted with a linear latitudinal scale and a linear barometric pressure scale. Figure 31 is plotted with a linear altitude scale. Several features of note may be seen in the several figures. First, the equatorial maximum appears to be fixed in location

*This value is obtained from the tungsten and strontium values averaged in the core of the September 1958 tropical concentrations. The tungsten value selected is 31,000 dpm and the strontium value selected is 400 dpm.

showing no significant vertical motion as would be expected in "Brewer-Dobson" circulation[15] or in the type of circulation suggested by Libby and Palmer.[16] Indeed, the HASP tungsten data are entirely incompatible with this latter suggestion although the general conclusions on the variability of residence time with altitude, latitude, and seasonal time of stratospheric injection reached by Libby and Palmer do agree with the conclusions to be drawn from the HASP data.[2,7] Second, the tungsten appears to spread northward and southward from the equatorial maximum along sloping lines showing a drop of about 15,000 feet by the time the debris reaches 70° latitude. Third, inventories calculated from these figures show that the time required for the first half of the tungsten to depart from the stratosphere is less than one year and that departure from the stratosphere is not exponential in time. Rather, as the debris ages, the apparent residence time increases. Fourth, by December 1959, the equatorial W-185 Sr-90 ratio (corrected to 15 August 1958) has dropped from the 85 figure previously noted to around 30. This indicates either that the tungsten has been leaving the stratosphere faster than the strontium or that the mixture has been diluted with extra strontium-90. The former conclusion seems more likely in view of the fact that the average HARDTACK-GRAPPLE Sr-90 was injected at higher altitudes than the tungsten. If there had been a substantial incursion of Soviet Sr-90, there would have been a drop in apparent age of the debris above the tropical tropopause. This has not been noted (see Chapter VI). Fifth, the concentrations in the Northern Hemisphere are greater than those of the Southern Hemisphere. The 4 to 1 ratio suggested by Lockhart[14] for January 1959 is about correct, however, the values have tended to equalize by May 1960. Finally, the tungsten values show a minimum in the vicinity of the tropopause gap. This could be caused by a large scale horizontal meandering flow of air back and forth between


MEAN DISTRIBUTION OF
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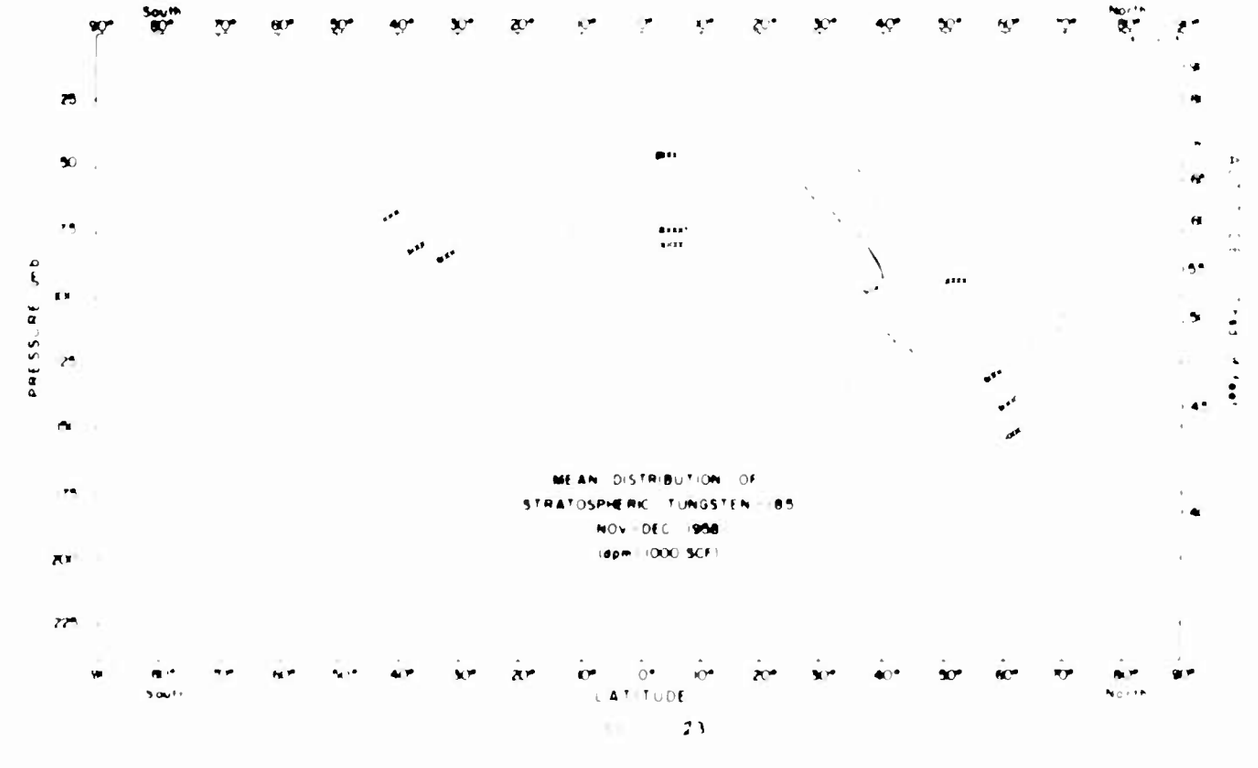
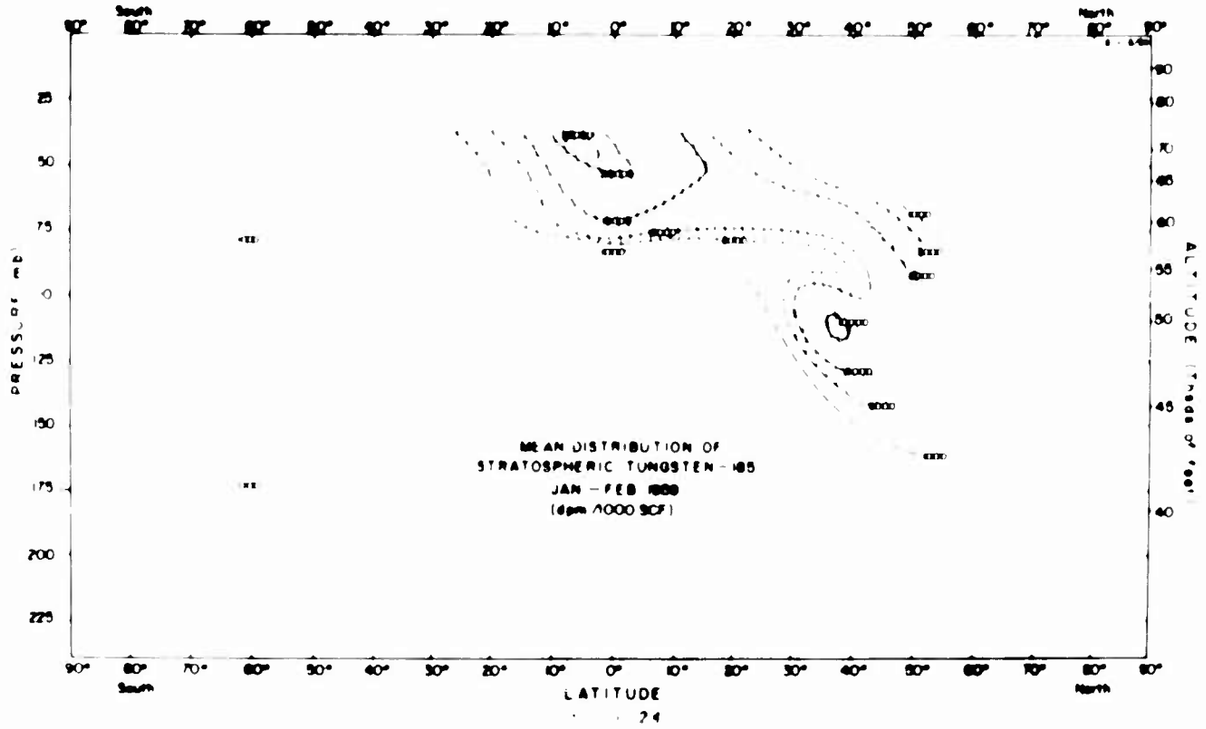
SEP - OCT 1958
(dpm/1000 SCF)
PRESSURE (MB)
LATITUDE
South
North

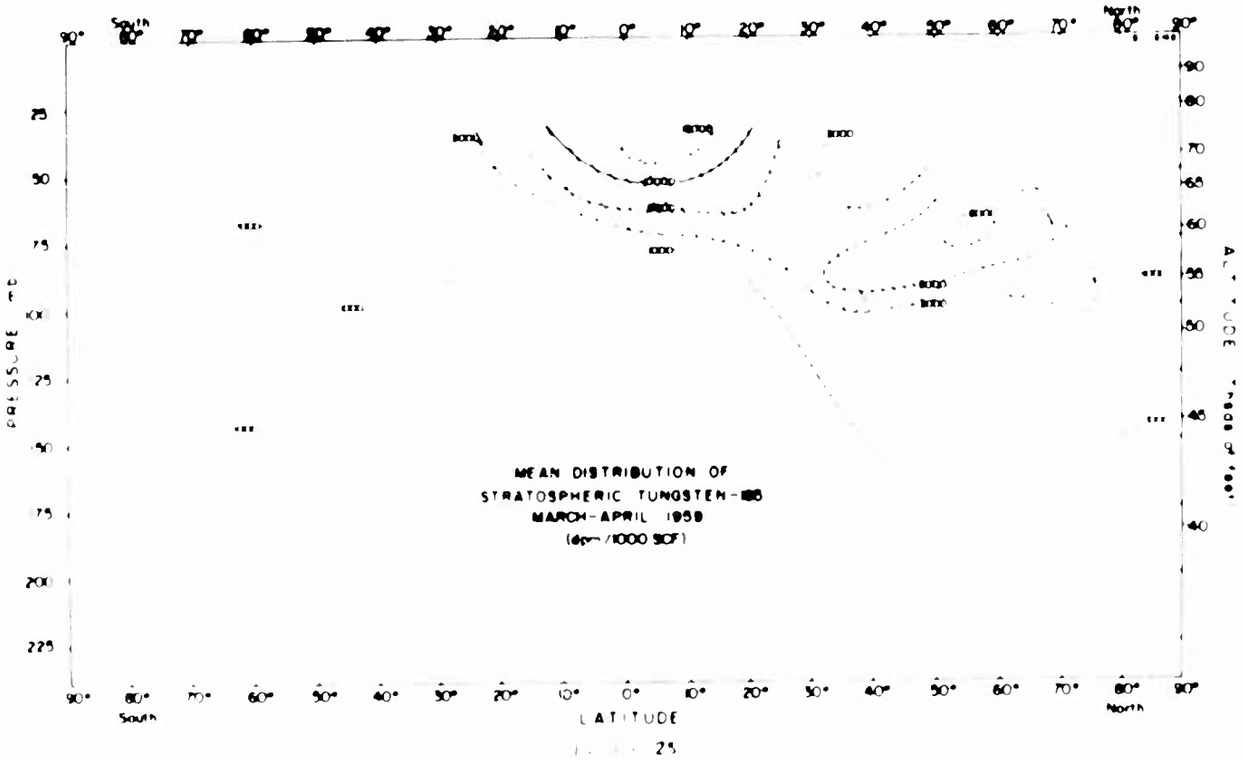
22


MEAN DISTRIBUTION OF
STRATOSPHERIC TUNGSTEN-185
NOV - DEC 1958
(dpm/1000 SCF)
PRESSURE (MB)
LATITUDE
South
North

23


MEAN DISTRIBUTION OF
STRATOSPHERIC TUNGSTEN-185
JAN – FEB 1959
(dpm/1000 SCF)
LATITUDE

24


MEAN DISTRIBUTION OF
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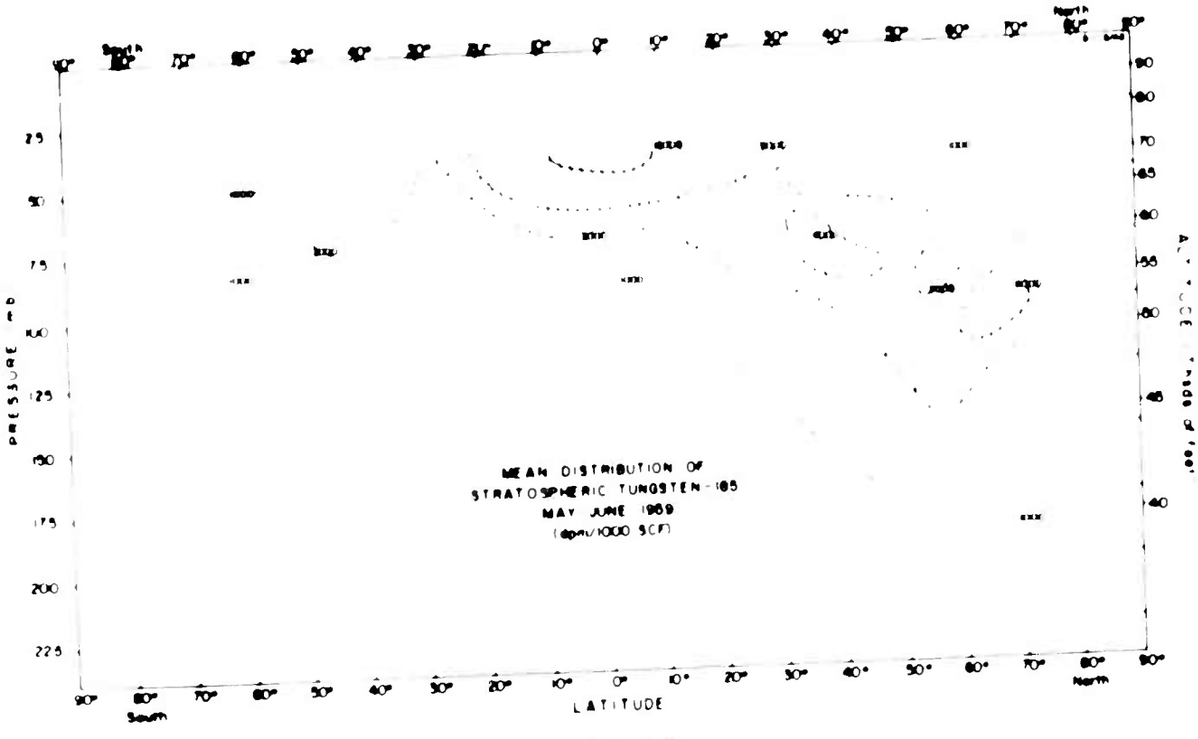
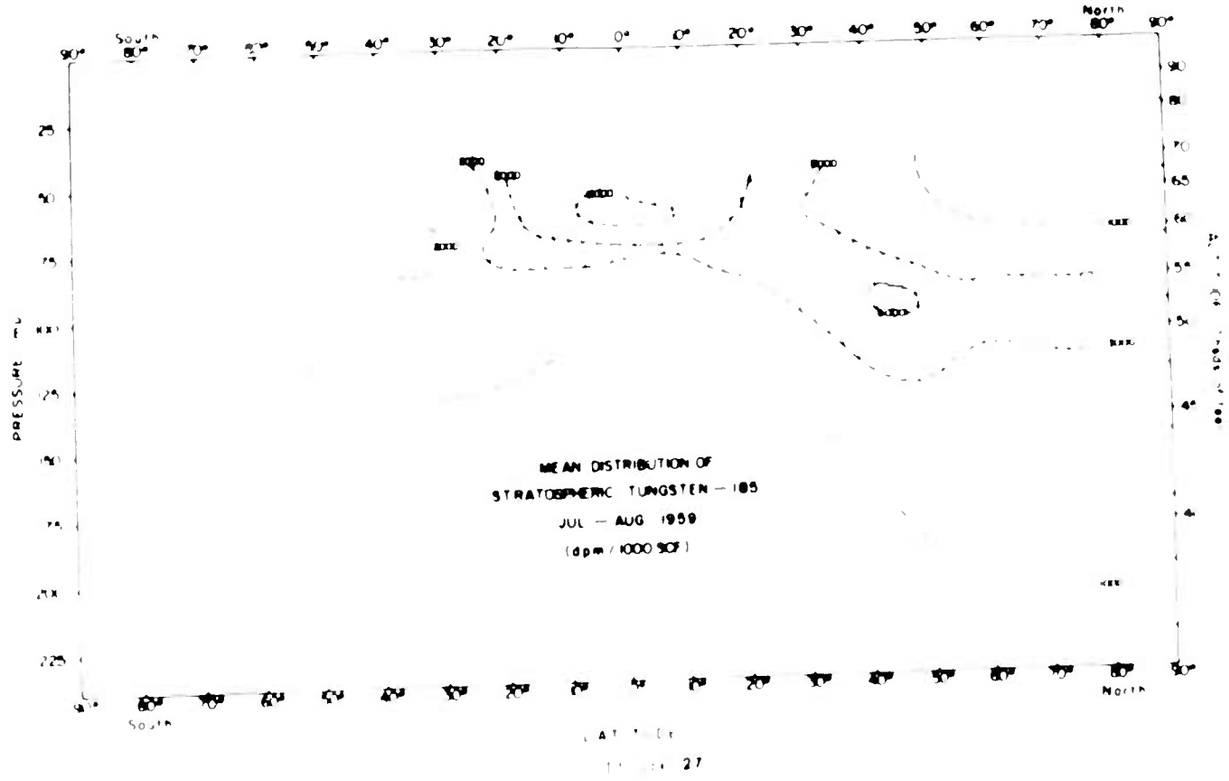
MARCH-APRIL 1959
(dpm/1000 SCF)
LATITUDE

25


MEAN DISTRIBUTION OF
STRATOSPHERIC TUNGSTEN-185
MAY JUNE 1959
(dpm/1000 SCF)
LATITUDE
South
North


MEAN DISTRIBUTION OF
JUL — AUG 1959
(dpm / 1000 SCF)
South
North

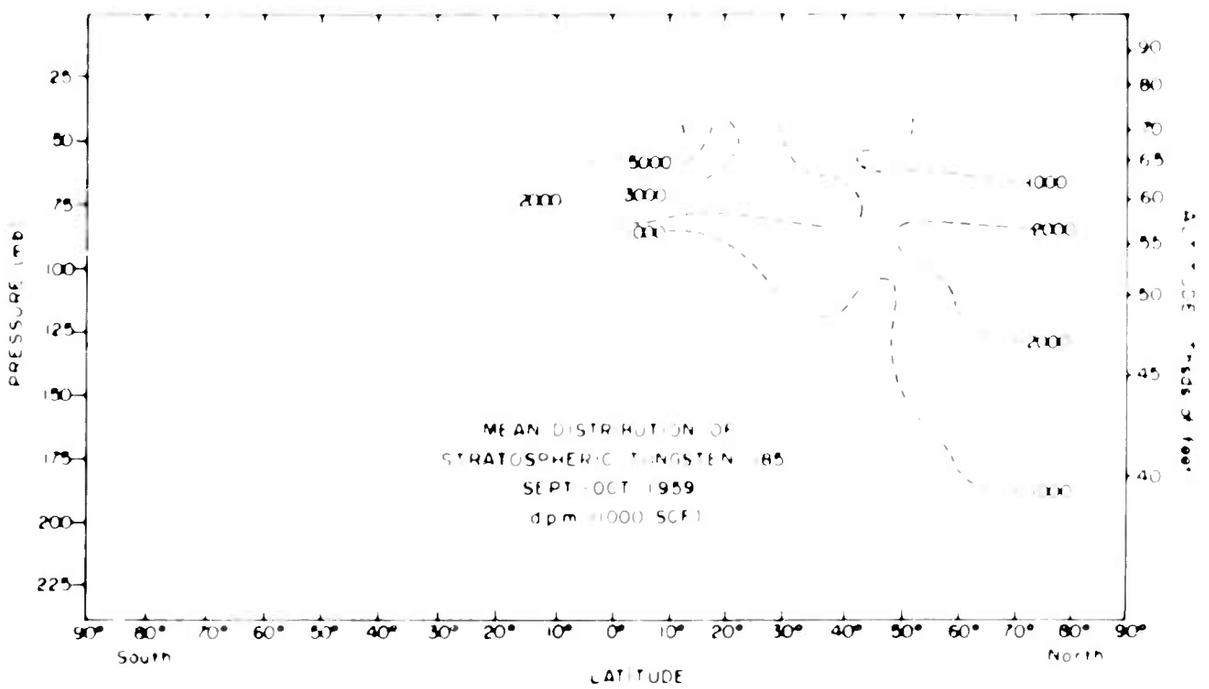


FIGURE 28

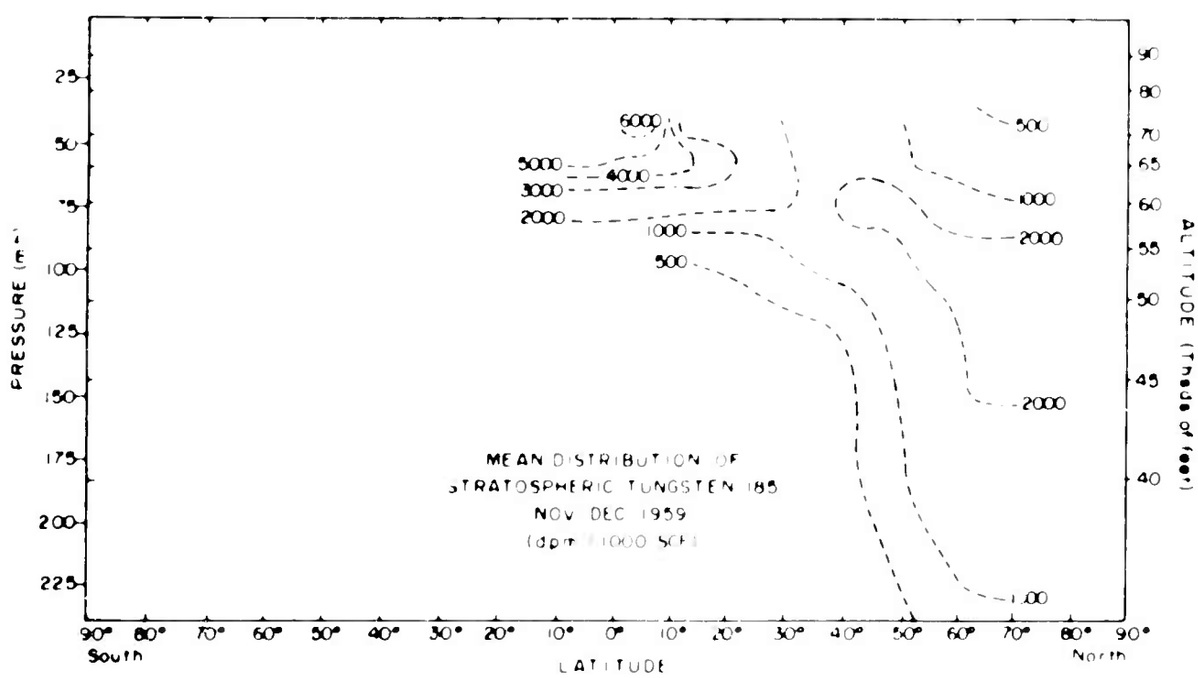


FIGURE 29

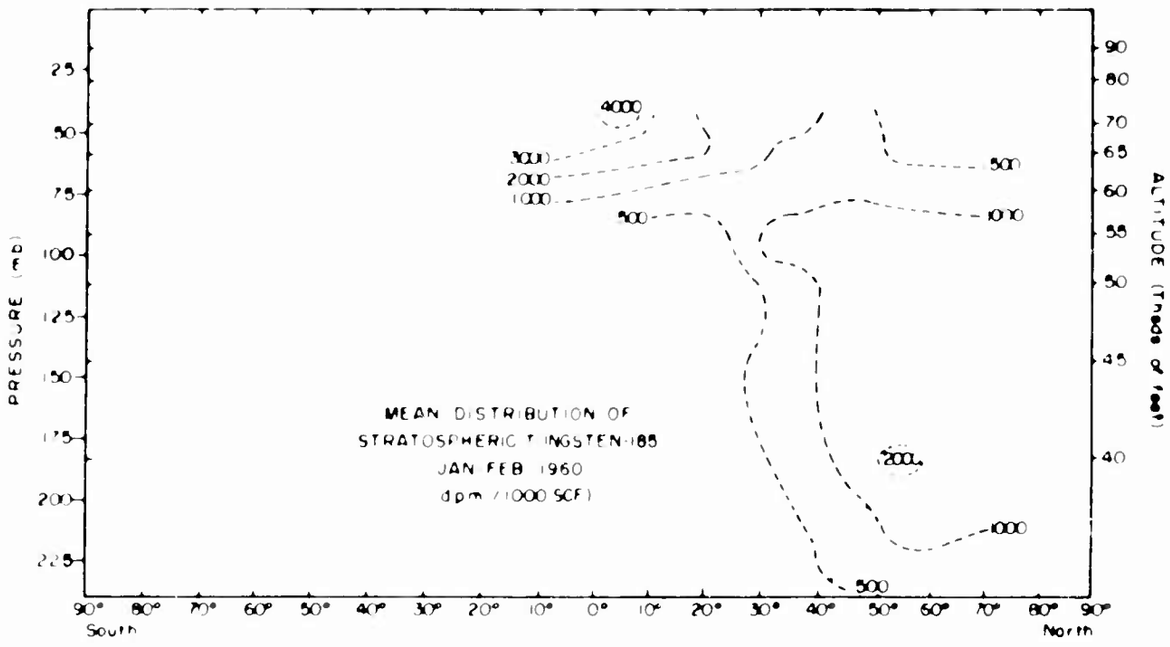
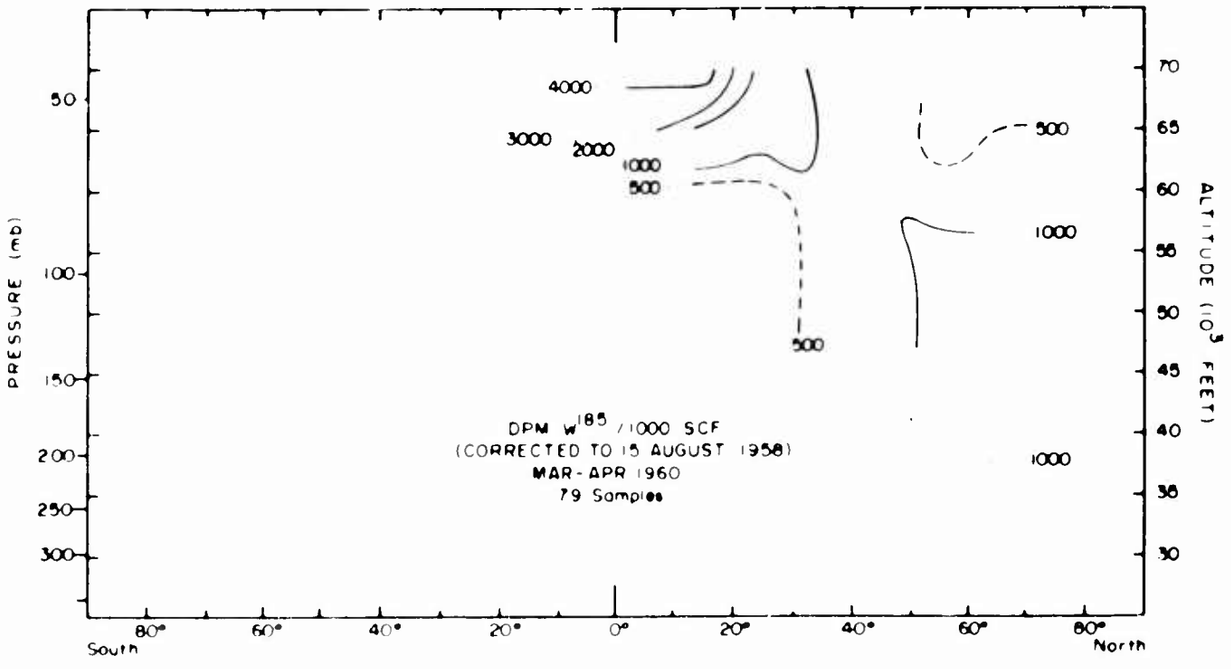


FIGURE 30



MEAN DISTRIBUTION OF TUNGSTEN-185, MARCH-APRIL 1960

FIGURE 31

the stratosphere and troposphere. This type of movement would cause material mixing poleward across the gap from the higher tropical regions to be placed alternately in the troposphere and in the polar stratosphere.

Some of the above phenomena are illustrated in Figure 32. This figure was constructed by selecting the maximum values of tungsten concentration within 5 degree latitude bands on a bimonthly schedule. Since the maxima slope downward toward the poles, the altitude represented at higher latitudes is progressively lower. The "wings" on the shoulders of the "normal" curve are quite evident, as is the gradual diminution of all the maxima with time.

All of the above tungsten observations can best be explained by the Austausch or turbulent diffusion concept of Spar[2,8,9] modified by the perturbations introduced by (1) seasonal changes in diffusion rates, (2) seasonal variations in the tropopause height, (3) seasonal variation in location and intensity of the jet stream, (4) seasonal variation in the location and structure of the tropopause gap. Further discussion of this material will be found in Chapter IX.

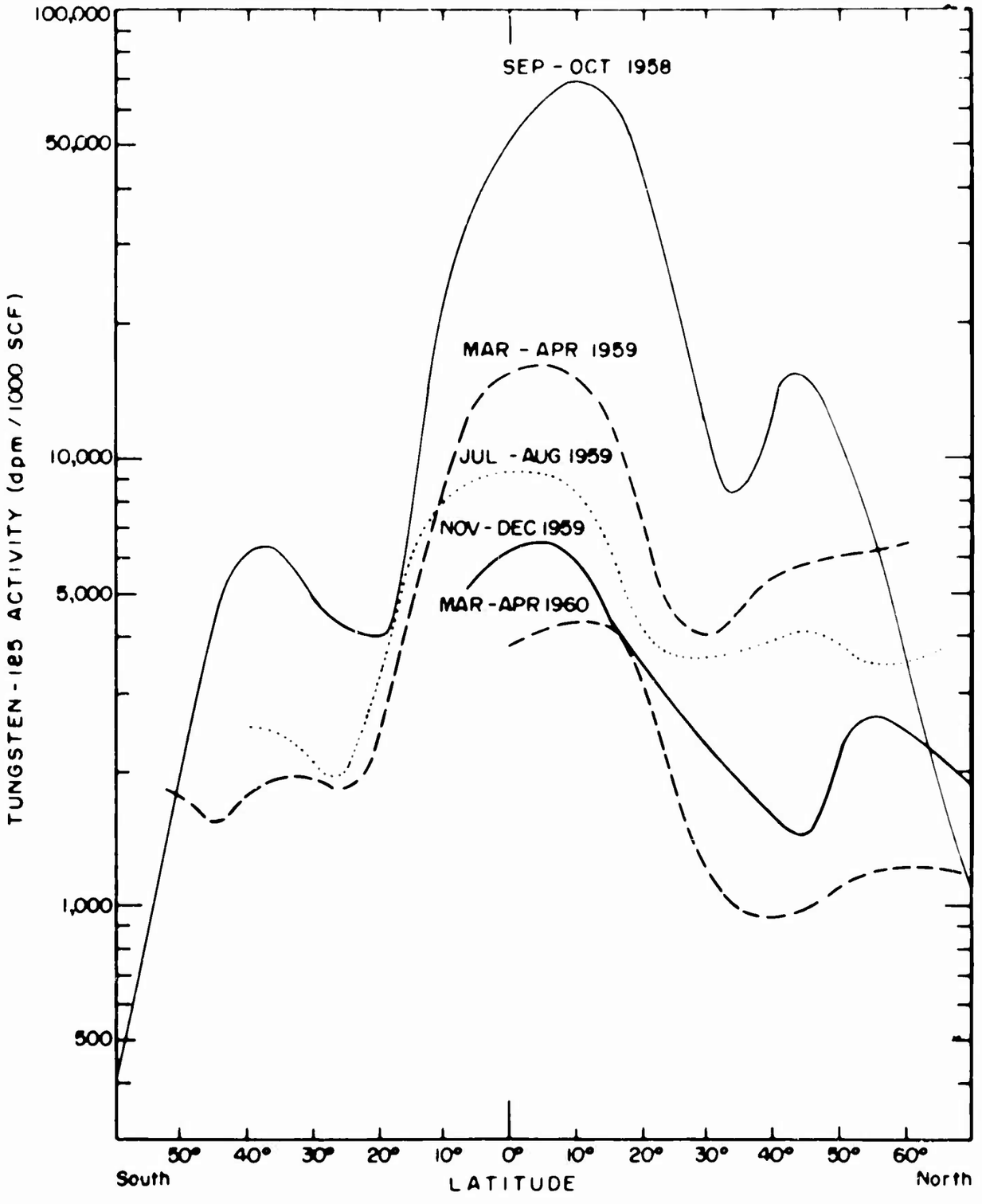


FIGURE 32 TUNGSTEN-185 ACTIVITIES (Corrected to 15 AUG 1958) IN THE ZONE OF MAXIMUM TUNGSTEN-185 CONCENTRATION

Chapter VI

BARIUM-140, STRONTIUM-89, CERIUM-144 RHODIUM-102 AND PLUTONIUM IN THE STRATOSPHERE

Introduction

When a number of atoms of fissionable material undergo the fission process, a large family of products is formed. Perhaps as many as 400 different radioisotopes with half lives varying from less than a second to more than 1000 years are formed. Dolan[35] has calculated the number of each of these elements to be expected as the result of simultaneous fast neutron (~14 Mev) fission of uranium-238. Ninety or more mass chains are produced, each containing a group of elements which more or less rapidly decay until only the element in the mass chain with the longest half life (plus its daughters) remains. After reaching this final stage, each isotope decays exponentially with a characteristic half-life according to the expression:

$$N = N_o e^{-\lambda t} \tag{3}$$

where $\lambda = 1/\tau = .693/T_{\frac{1}{2}}$ (4)

N being the number of atoms remaining at time, t, after the initial (or apparent initial) quantity, N_o, was available. λ is the decay constant characteristic of each element and is related to the mean life, τ, and half-life, $T_{\frac{1}{2}}$, as indicated in (4). The activity or rate of decay at any time, t, is given by a similar expression.

$$dN/dt = A = A_o e^{-\lambda t} = N\lambda = -N_o \lambda e^{-\lambda t} \tag{5}$$

To a fair approximation, mixed fission products diminish in activity according to the relationship:

$$A = A_1 t^{-1.2} \tag{6}$$

where A_1 is the activity at unit time after fission and A is the activity at some time, t, later (or earlier).

Table VIII is a tabulation from Dolan's report of all fission products with a half-life greater than 5 days. These elements were selected to illustrate the possible radioelements available for study in stratospheric or long range tropospheric fallout. They are arranged in order of increasing half-life. Except for mass number 147, the elements shown are those in any one mass chain which have the longest half-life. The missing mass chains between 85 and 161 have no members with half-life greater than 5 days. With the exception of Sb-125, Te-127*, I-131, Xe-133, Pr-143, and Pm-147, the precursors of the elements shown essentially will have all decayed within one month after detonation. In the case of the exceptions listed above, less than 6% of the precursors will remain one month after detonation.

Normally samples collected in the HASP network have not been processed in the laboratory in a period of time less than one month after detonation of the weapon represented in the filter. This appears to be the case in the other collection programs referred to herein. The half-lives shown in the table are expressed in days and the production number is that number of atoms of the material in question which apparently is produced for every 10,000 fissions at the instant of detonation when observed after all the precursors have decayed. This number is slightly greater than the sum of the atoms of the precursors and the element in question at the instant of fission due to the holdup time of final production concomitant with the decay of the precursors.

*The asterisk indicates an isomer of the nuclide

Table VIII

Fast Fission Product Production in U-238

Element & Mass No	Half-Life	Production	Element & Mass No	Half-Life	Production
Xe-133	5.27 d	610	Ce-141	285 d	330
Tb-161	6.88 d	1	Ru-106	369 d	270
Ag-111	7.6 d	81	Eu-155	620 d	25
I-131	8.1 d	130	Sb-125	730 d	89
Nd-147	11.1 d	220	Pm-147	65 d	222
Ba-140	12.8 d	156	Kr-85	3760 d	30
Cs-136	12.9 d	3	Cs-137	9710 d	521
Ir-143	13.76 d	396	Sr-90	10100 d	300
Eu-156	15.1 d	16	Sm-151	31000 d	105
Ce-141	33.1 d	432	Be-7	53 d	0
Te-129*	33.5 d	61	C-14	5760 y	0
Ru-103	39.8 d	450	P-32	14.5 d	0
Cd-115*	42.6 d	6	Rh-102	210 d	0
Sr-89	50.5 d	240	Rh-102*	?	0
Y-91	57 d	344	W-181	120 d	0
Zr-95	65 d	470	W-185	75.8 d	0
Te-127*	105 d	30	Pb-210	8030 d	0
Sn-123	136 d	78	Pu-239	24,300 y	0

Note: The elements with zero production are listed here for convenience only.

A number of these isotopes are not suitable for study of fallout debris for one reason or another. The noble gases cannot be collected by a particulate filter, and some elements are volatile and will not remain in the filter paper residue during the ashing process prior to radiochemical separation. The decay schemes of some elements are such that they are not amenable to accurate assay either because the schemes are not presently well known or because the beta or gamma emission spectra of several elements overlap. Some elements such as Cd-115*, Cs-136, and Tb-161 are not produced in great quantity as are other more tractable elements.

As far as studies of world-wide fallout are concerned, only about a dozen mass chains are worthy of study. Sr-90 and Cs-137 are studied because their half-lives are long compared to stratospheric residence times and they are therefore useful in measuring the stratospheric burden and mixing rates. They also are major contributors to the long term fallout hazard. Zr-95, Ru-103, 106, Ce-111, 141 are studied because they, along with Cs-137, are the major sources of external gamma radiation in world-wide fallout. Ce-141 is also useful for dating older debris. Sr-89 and Y-91 are useful for dating debris less than one year old and Ba-140 is useful for dating debris only a few months old. Mixed fission product beta half-life is another useful but perhaps less accurate method of dating debris up to a few months old. I-131 may be an important radioelement to consider in tropospheric fallout due to its strong tendency to concentrate in the thyroid gland. Other natural or neutron activation products which are worthy of study for the light they shed on stratospheric mixing processes or their contribution to the fallout hazard include C-11, Pu, W-181, 185, Pb-210, Rh-102, P-32, and Be-7.

Dating Radioactive Debris

One method of determining the age of unmixed, unfractionated radioactive debris is to measure the ratio of activities of two isotopes in the sample. If the initial activity and half-life of each are known, the ratio can be expressed as shown below.

$$R = \frac{A_a}{A_b} = \frac{A_{O_a} e^{-\lambda_a t}}{A_{O_b} e^{-\lambda_b t}} = \frac{N_{O_a} \lambda_a}{N_{O_b} \lambda_b} e^{-(\lambda_a - \lambda_b)t} = R_O e^{-\lambda_r t} \quad (7)$$

where R represents the ratio of the isotopic activities, A, (say, in dpm) of elements a and b at some time, t, after detonation. It is customary to form the ratio by dividing the activity of the element with the shorter half-life by the activity of the element with the longer half-life. It can be seen that the ratio decreases from some initial value

$$R_O = \frac{N_{O_a} \lambda_a}{N_{O_b} \lambda_b} \quad (8)$$

with a characteristic decay constant

$$\lambda_r = \lambda_a - \lambda_b \quad (9)$$

The half-life of the ratio is given by the expression:

$$T_{\frac{1}{2}r} = T_{\frac{1}{2}b} \times T_{\frac{1}{2}a} / (T_{\frac{1}{2}b} - T_{\frac{1}{2}a}) \quad (10)$$

Knowing the ratio at the time of analysis one can calculate the time elapsed since the ratio was at its original value. This then is the age of the debris. In principle, measurement of the activity ratios from any two penultimate elements from different mass chains should produce the same age. As we shall see below, several complicating factors can arise which obscure this picture.

Using the values of N_O and $T_{\frac{1}{2}}$ shown in Table VIII one can determine the initial activity ratios, R_O, and apparent ratio half-life $T_{\frac{1}{2}r}$ for a number of nuclides.

Table IX shows these values for the more important isotopes mentioned above. The upper and lower figures at the row and column intersection of any two elements are, respectively, R_0 and $T_{\frac{1}{2}r}$ expressed in days. For example, the Sr-89/Sr-90 activity ratio at t = 0 apparently is 160 and it decays with a half life of 50.7 days. Although W-185 is not a fission product, it is included in the table for convenience. The two values of R_0 shown in each box are based on the estimated tungsten to fission yield values for the HARDTACK and HARDTACK-[illegible]PLE series respectively with t_0 = 15 August 1958. These are the stratospheric injection values discussed in Chapter V.

If unfractionated clouds of two similar weapons exploded on different dates mix together, it is possible, in principle, to determine the dates of explosion of each and the relative fraction of the debris from each in the sample collected. This can be done by making a judicious selection of several isotopes for activity measurement. In practice, however, this becomes quite difficult and may even be impossible. The difficulties have been discussed extensively by Lockhart[31,33] and include: (1) fractionation of one isotope with respect to another during transport through the atmosphere, (2) fractionation by the collection system, (3) use of different fissionable materials subjected to a variety of neutron energy spectra resulting in different values for R_0, (4) upward shift of the fission product mass spectrum by multiple (n,γ) reactions on the fission products, (5) different times of arrival of debris from the same event due to varied paths of travel, (6) uncertainties in the measurement of isotopic concentrations in the samples collected from uncertainties in decay schemes, low levels of activity or masking by other elements, (7) a background residuum left by a series of earlier bursts, and (8) the influx of material from a non-weapon source such as the I-131 which

Table IX

Fast Fission Product Isotopic Activity Ratio

Ba^{140}	2.73 20.9d	3.15 18.9d	7.50 17.6d	5.90 16.5d	4.93 15.9d	5.55/14.1 15.4d	30.7 13.4d	48.1 13.3d	661 12.8d	1200 12.8d
	Ce^{141}	1.155 164d	2.75 96d	2.16 79d	1.81 67.5d	2.04/5.18 58.8d	11.25 36.5d	17.65 36.3d	243 33.2d	440 33.2d
		Ru^{103}	2.38 188d	1.87 132d	1.56 103d	1.76/4.47 83.8d	9.75 44.7d	15.3 44.6d	210 40d	381 40d
			Sr^{89}	0.79 443d	0.656 226d	0.74/1.88 153d	4.1 61.3d	6.42 58.5d	88.5 50.7d	160 50.7d
				Y^{91}	0.835 463d	0.94/2.49 230d	5.2 71.3d	8.15 67.4d	112 57.3d	203 57.3d
					Zr^{95}	1.13/2.87 456d	6.24 84.2d	9.78 73.9d	134 65.4d	244 65.4d
						W^{185}	5.53/2.18 103d	8.67/3.41 95.4d	119/48 76.3d	216/85 76.3d
							Ce^{144}	1.57 1250d	21.5 293d	39 293d
								Ru^{106}	13.8 383d	24.9 383d
									Cs^{137}	1.82 244,000d
										Sr^{90}

escaped into the atmosphere in the Windscale incident. Surface samples measuring total activity taken near a reactor site are especially subject to this latter source of error.(36)

Another method of determining the age of unmixed, simultaneously fissioned debris is to determine the apparent half-life of the total beta (or gamma) emission from the sample. If it is assumed that the $t^{-1.2}$ law holds, then the apparent decay constant, λ, at any one moment after detonation is a decreasing function of the time since burst. This value of λ can be determined by constructing an exponential activity curve which has the same value and same slope as the actual activity curve at some selected time. Differentiating equations (5) and (6) one obtains:

$$\frac{dA}{dt} = -\lambda A_0 e^{-\lambda t} = -\lambda A \qquad \text{and} \qquad (11)$$

$$\frac{dA}{dt} = -1.2\, A_1 t^{-1.2} \times t^{-1} = -1.2\, At^{-1} = \lambda A \qquad (12)$$

Solving for the apparent age, t, one obtains finally:

$$t = 1.2/\lambda = 1.2\tau = 1.73T_{\frac{1}{2}} = T_{.3} \qquad (13)$$

where $T_{.3}$ is the apparent time required for the material to have its activity reduced to three-tenths of its original value (alternatively the apparent time required to lose 70% of its activity). It should be noted that the apparent times mentioned above refer to the mathematically constructed exponential curve used to determine the age and are not the actual times required for the activity of the real debris to diminish by a certain amount, since these, of course, increase as the age increases. A simple method of determining the age using this technique is to plot the activity of the sample on semi-log paper and to draw a tangent to the curve at the selected date. The time required for this tangent to drop from 10

to 3 is, then, the age of the debris at the point in time at which the tangent is drawn.

Table X shows some typical values of fresh radioisotopes intercepted in the HASP network during 1958. In addition, a sample of French debris collected by B-57 aircraft during the spring of 1960[37] is included. All the data are expressed in dpm/1000 scf corrected to sampling date except that the tungsten is corrected to 15 August 1958. The beta half life is that value observed 10 days after the date of collection.

It can be seen that the fresh fraction of debris in the atmosphere found in the presence of older debris can be dated quite well using either Ba-140/Sr-89 ratios (assuming all of both isotopes came from the most recent shot) or beta half-life. Samples 1, 2, 6 and 7 fairly accurately date debris from USSR tests as do samples 3 and 5 for UK debris. Sample 3 is probably of tropospheric origin. Sample 4 from the United States test shows evidence either of some fractionation or of mixing with the earlier UK shot, since the shot date is not pinpointed by the beta age as well as the others.* The Sr-89/Sr-90 ratio does pinpoint the date fairly well, however. Sample 4 has a great deal of tungsten-185 in it with a W-185/Sr-90 ratio at shot time of at least 1450. This is the same order of magnitude for the maximum ratio calculated by Lockhart[31] from data collected along the 80° meridian in June 1958.

In general, the apparent age calculated using ratios with the strontium-90 in these samples will yield an earlier date of apparent origin than that obtained with the nuclides of shorter half-life. However, if the background values of Ce-144, Sr-90, Sr-89 and Zr-95 observed in samples obtained during the month prior to the fresh intercept are subtracted from the values shown in the table, then most of the ratios generally tend to approach their respective theoretical values. In the

* The presence of large quantities of activated tungsten will tend to increase the apparent age based on beta half li. of mixed fission products.

TABLE X

Radioactivity in Fresh Debris

	Sample No	Collection Date	Latitude	Altitude	dpm/1000 scf *					
					Ba-140	Sr-89	Zr-95	Ce-144	Sr-90	*-185
1.	329	25 Mar 58	30° N	60 K	212 K	78 K	104 K	28 K	856	---
2.	408	8 Apr 58	53° N	55 K	48 K	13.6K	15.5 K	---	174	---
3.	488	6 May 58	0° N	55 K	---	180	---	---	1.3	---
4.	563	24 June 58	8° N	60 K	4.4 K	10 K	5.78 K	---	114	110 K
5.	654	3 Oct 58	1° N	60 K	275 K	118 K	151 K	38.8 K	1080	31.5 K
6.	732	22 Oct 58	44° N	60 K	162 K	47 K	69.7 K	---	936	5.8 K
7.	751	29 Oct 58	41° N	60 K	275 K	145 K	133 K	41 K	1683	3.02 K
8.	023	8 Mar 60	35° N	42 K	27.6	14.5	12.2	---	17.8	520

	Sample No	Beta Half Life	Beta Age	Ba-140/Sr-89	140/89 Age	Apparent Source	
1.	329	22 d	25 Feb 58	2.72	27 Feb 58	27 Feb 58	USSR
2.	408	22 d	1 Mar 58	3.53	20 Mar 58	21 Mar 58	USSR
3.	488	12 d	25 Apr 58	---	29 Apr 58 **	28 Apr 58	UK
4.	563	43 d	21 Apr 58	0.44	18 Apr 58	11 or 12 May 58	US
5.	654	22 d	5 Sep 58	2.33	3 Sep 58	2 Sep 58	UK
6.	732	17 d	2 Oct 58	3.45	2 Oct 58	2 Oct 58	USSR
7.	751	21 d	3 Oct 58	1.90	23 Sep 58	2 Oct 58	USSR
8.	023	---	---	2.52	3 Feb 60	12 Feb 60	French

* Corrected to sampling date except *-185 which is corrected to 15 Aug 1958.
** From Sr-89/Sr-90 ratio

case of weapons fired well above the surface of the earth (such as in the case of the 1958 US and USSR detonations) little fractionation is noted. Surface bursts will display a greater degree of fractionation. For instance, the equatorial samples collected from 1958 through 1960 seem to be depleted in Ce-141.

Distribution of Barium-140 and Strontium-89

As described in the previous section, barium-140 and strontium-89 can be used to date debris a few months old. As a consequence, a study of the ratio of these isotopes from place to place immediately after a test series can shed some light on the extent of mixing in the stratosphere during the first few months after the series. Figure 33 displays the Ba-140/Sr-89 ratios in the stratosphere during late 1958. The dotted lines show the theoretical slope from the UK GRAPPLE shots and Soviet series in September and October 1958. Different latitude bands are represented by the shape of the plotted points.

In the 30° to 10°S band, the ratio has risen by early October from a low value to that expected from the UK shots but then drops away faster than the theoretical value. This fall is probably due to an incursion of older Sr-89 from the HARDTACK series. In the equatorial belt the ratio rises to the value expected from the UK shots by the end of September and falls along the theoretical slope until mid-November when the ratio suddenly rises to the value expected from the Soviet test series. The time lag noted between the UK shots and the peak in the ratio is a measure of the time required for the easterly zonal winds to carry the debris from the test site into the HASP sampling network. The mid-November rise is apparently due to the southward mixing of the Soviet debris.

The debris noted in the 20° N to 30° N band represents a mixture of Soviet and UK debris. The UK debris did not contribute as much here as it did in the equatorial

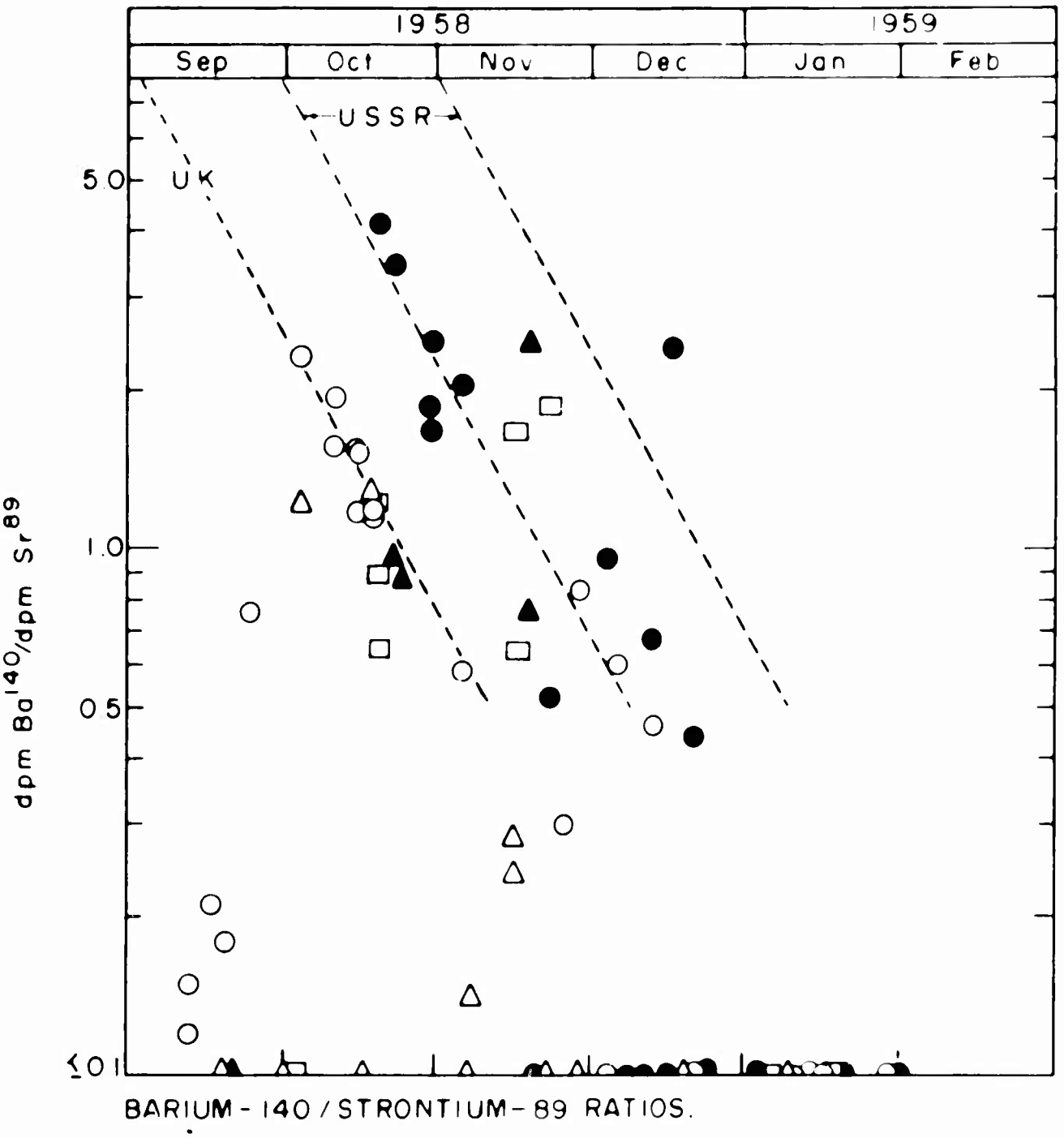

1958
1959
Sep
Oct
Nov
Dec
Jan
Feb
USSR
UK
5.0
1.0
0.5
≤0.1
dpm Ba140/dpm Sr89
BARIUM - 140 / STRONTIUM - 89 RATIOS.
30°S - 10°S
10°S - 20°N
20°N - 30°N
30°N - 50°N
50°N - 70°N

FIGURE 33

region (the latitude of the test) while the Soviet debris contributed more here than it did in the equatorial region. One would expect the high latitude Soviet debris which reached the equatorial region to have passed through the temperate region. This is shown to be so by these data since the temperate zone values show proportionately more debris than the equatorial region and also show an earlier rise.

In general, the ratios noted in the 30^{O} N to 50^{O} N band show Ba-140 and Sr-89 exclusively from the Soviet shots of early October. However, the debris measured at 50^{O} N to 70^{O} N (the latitude of the Soviet test site) indicates somewhat lower values. Apparently the circulation in the polar stratosphere was not symmetrical around the pole at this time but circulated around a point displaced toward Canada. As a result, older Sr-89 from the HARDTACK and GRAPPLE tests depressed the values in the highest latitude region.

Since the half-life of Sr-89 is about three times that of Ba-140, Sr-89/Sr-90 ratios can usefully trace debris from various nuclear test series up to a year after the series. In addition, some of the tendency for fractionation between isotopes is reduced since Sr-89 and Sr-90 are chemically similar and both have a rare gas precursor. Figure 31 shows Sr-89/Sr-90 ratios plotted versus time for three regions of the stratosphere: the tropical stratosphere (30^{O} North - 30^{O} South), the northern polar stratosphere (north of 30^{O} North) and the southern polar stratosphere (south of 30^{O} South). For the tropical stratosphere only samples collected at 60,000 to 70,000 feet were used. For the northern polar and southern polar stratosphere the data were divided into three sets: one for samples collected at 30,000 to 35,000 feet, a second for samples collected at 40,000 to 55,000 feet, and a third for samples collected at 60,000 to 70,000 feet. A curve has been drawn through the points for the tropical stratosphere and this curve has been repeated in the other two regions for purposes of comparison.

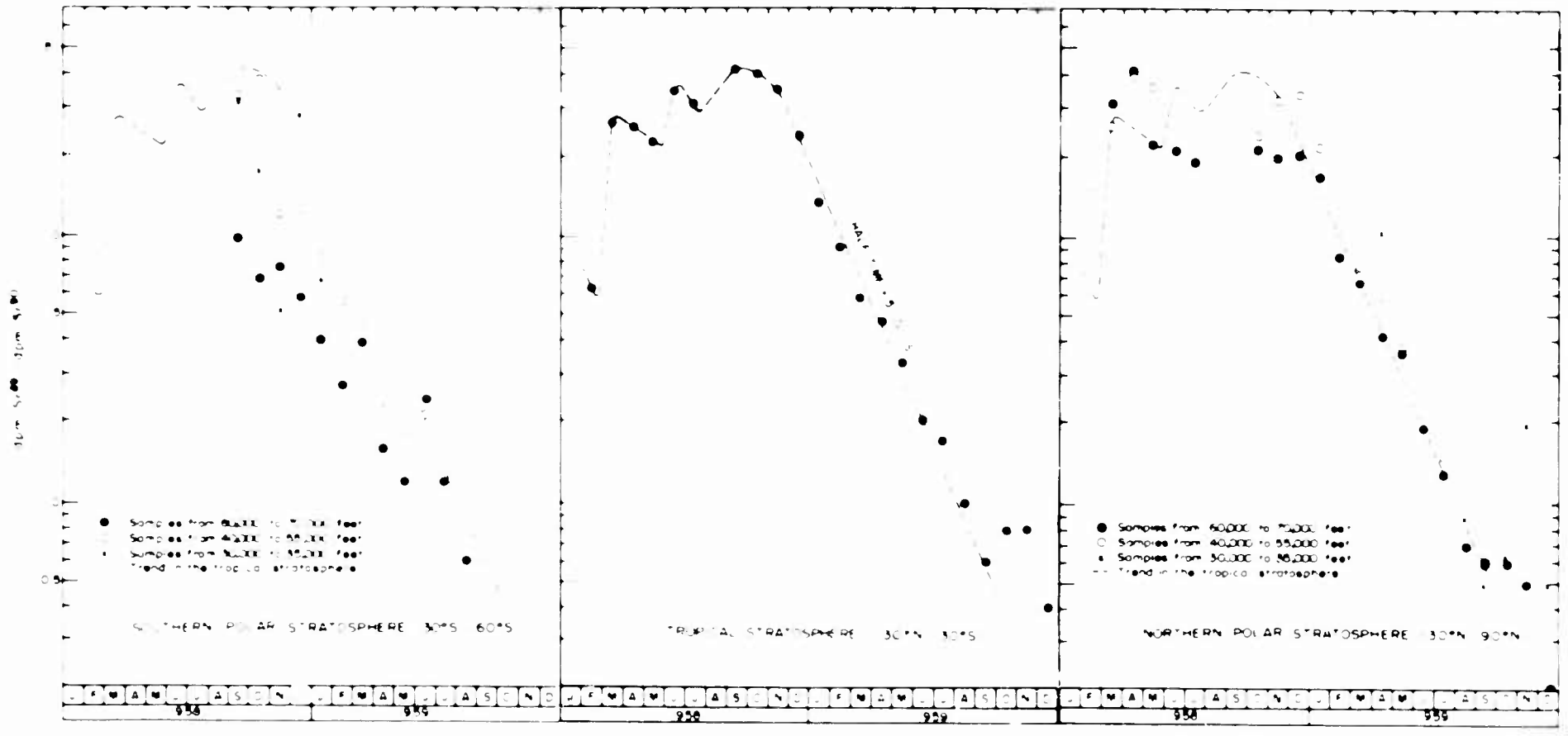


FIGURE 34 VARIATION WITH TIME OF THE MONTHLY AVERAGE FOR Sr^{89}/Sr^{90} SAMPLES

Between November 1958 and September 1959 the ratio in debris in the tropical stratosphere fell in response to the decay of strontium-89. The apparent date of origin of this debris, obtained by extrapolating the line back to a ratio of 160, is 25 July 1958, in good agreement with its derivation from HARDTACK. After September 1959 the ratio ceased to fall, either because of the influx of younger material from some other region of the stratosphere or, more likely, because the strontium-89 could not be measured accurately in the low concentrations present in the tropical stratosphere in late 1959.

By January 1959, the debris sampled in the northern polar stratosphere had the same apparent age as that sampled in the tropical stratosphere. This correspondence in apparent age between the polar and tropical stratosphere continued until at least September 1959.

There are two explanations for the apparent age of the debris in the northern polar stratosphere. The first is that it represents a mixture of debris from the October 1958 Soviet tests with older debris. If it is assumed that the older debris contained no strontium-89, the Soviet debris contributed about 27 percent of the strontium-90 in the polar stratosphere. If the old debris did contain strontium-89, less than 27 percent of the strontium-90 came from the October 1958 Soviet tests. The second explanation for the apparent age assumes that virtually all debris from Soviet tests had fallen out, or at least had not been mixed into the part of the stratosphere which HASP was sampling, and that the apparent age of the debris was caused by the transfer of large quantities of debris from HARDTACK into the polar stratosphere. Perhaps some Soviet debris remained in the very low stratosphere below 40,000 feet but, if it did, it was certainly present even there only in low concentrations.

The change in Sr-89/Sr-90 ratio which occurred in the northern polar stratosphere in October 1959 is similar to that which occurred in the tropical stratosphere at the same time. Again it seems more likely that the failure of the ratio to continue falling is due to analytical error rather than to the influx of young material, especially since the ratio for debris from 40,000 to 55,000 feet became higher than that for debris from 60,000 to 70,000 feet and the only likely source of younger material lay above 70,000 feet.

Measurements of the Sr-89/Sr-90 ratio in surface air made by Lockhart[33] show that approximately 75 to 80 percent of the Sr-90 activity in the northern hemisphere during the spring of 1959 came from the Soviet series, with activities before February and after August showing higher concentrations of older debris (HARDTACK and GRAPPLE). The apparent disappearance of Soviet debris from the northern stratosphere supports the suggestion made by various investigators[33,39,40,41] that most of this debris was removed from the atmosphere during the spring of 1959 (See Chapter X).

The Sr-89/Sr-90 ratio in debris in the southern polar stratosphere was lower than that in debris in the tropical and northern polar stratosphere throughout early 1959. However, the ratio increased steadily during this interval and by mid-1959 it was the same (or almost the same) in the southern polar stratosphere as it was in the tropical stratosphere. Even in September and October 1958 the apparent age of debris in the Southern Hemisphere was only two months older, for debris at 40,000 to 55,000 feet, or five months older, for debris at 60,000 to 70,000 feet, than that of debris in the tropical stratosphere. The variation with altitude of the apparent age of this debris suggests that either HARDTACK debris mixed preferentially into the low southern polar stratosphere or that HARDTACK debris from higher altitudes mixed with more older debris (either in the tropical or in the southern polar

stratosphere) than did HARDTACK debris from lower altitudes.

Lockhart's[33] measurements of Sr-89 at the surface in the Southern Hemisphere show a trend of the Sr-89/Sr-90 ratio during 1959 similar to that observed at the 60,000 foot level with values slowly changing from about 1/5th that of the Northern Hemisphere in January to about equal amounts in both hemispheres by September.

Distribution of Cerium-141

As seen above, strontium-89 measurements in the stratosphere are useful in tracing debris for about one year before the concentrations become too low for accurate analysis. Cerium-141 with its half-life of 285 days may be useful for periods up to 3 or 4 years. Figure 35 shows the Ce-141/Sr-90 ratios measured in HASP and some Ashcan samples versus time for three regions of the stratosphere. The depiction is similar to that of the Sr-89/Sr-90 ratios shown in Figure 34.

For samples collected in the tropical stratosphere since September 1958, the data fit the theoretical decay curve for this ratio quite satisfactorily as did the Sr-89 data. For samples collected in the northern polar stratosphere during this same interval the data show more scatter and there is a distinct difference between the ratio in samples collected at 55,000 feet and below and that in samples collected at 60,000 feet and above. The data for samples collected in the southern polar stratosphere show even more scatter though there is no obvious altitude effect. In all regions of the stratosphere which were sampled, the apparent age of the debris is older according to this ratio than to the Sr-89/Sr-90 ratio, probably because the bulk of the debris present in these regions was derived from United States ground shots in the Pacific which produced fractionation of strontium-90 relative to cerium-141. Only two important sources exist which could contribute debris with a Ce-141/Sr-90 ratio higher than that found in debris in the tropical stratosphere;

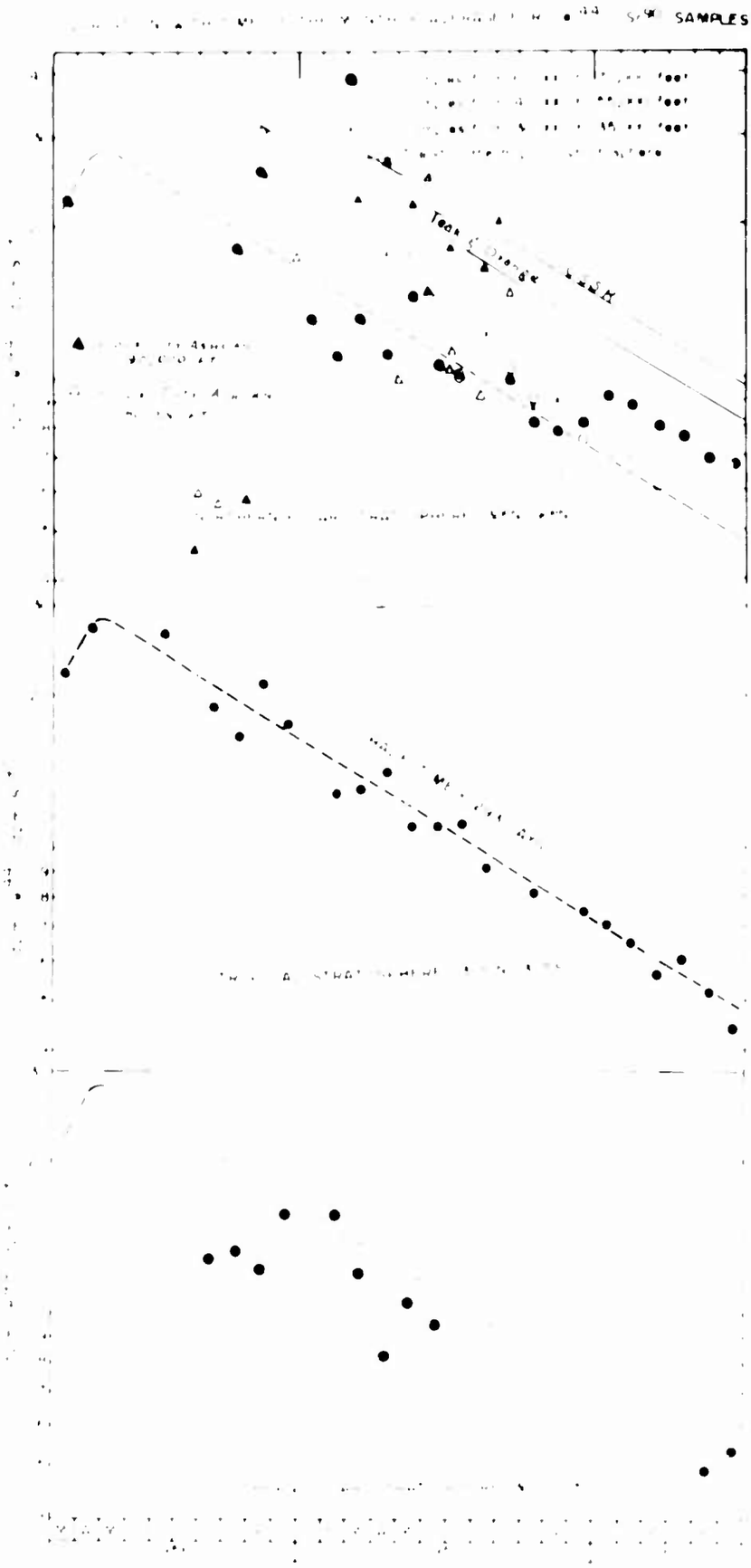


Figure 35

material from the October 1958 Soviet, and material from the August 1958 United States high altitude rocket shots. The ratio should be higher in debris from these sources since they probably produced no fractionation and were fired later. Figure 35 shows the theoretical slope from these two sources.

HASP Data

In the northern polar stratosphere during the spring and summer of 1959 there was a negative gradient in the Ce-144/Sr-90 ratio, the values at the highest altitude being lower than the tropical values while the values below 60,000 feet being higher than the tropical values. As time progressed, most of the younger debris at the lower altitudes (from the Soviet series) departed from the stratosphere while some of the remainder may have mixed upward. By October 1959 all altitudes had the same ratio as that of the tropical stratosphere. The debris present at 60,000 to 70,000 feet had a slightly lower ratio than found in the tropical stratosphere probably due to a small background of pre-HARDTACK debris. The negative gradient observed in these cerium-144 data is not as apparent in the strontium-90 data above. This is probably due to the fact that the Soviet debris which accounts for the higher values at lower altitudes is relatively unfractionated while the HARDTACK debris is relatively more depleted of cerium-144. In any event, the cerium-144 data suggests that the Soviet contribution during early 1959 accounted for no more than 20% of the Strontium-90 activity in the northern stratosphere below 80,000 feet.

Starting in October 1959 and concurrent with the rise in Sr-90 concentration noted in Figure 21, the Ce-144/Sr-90 ratio makes an abrupt rise at the 60,000-70,000 foot level. A few months later a similar rise is noted at the lower altitudes. It is quite evident that this material did not come from the tropical regions sampled. It is suggested that the major fraction of this increase is due to an incursion of

debris from Teak and Orange. Some quantitative estimate of the fraction of Teak and Orange in this region can now be made.

From Figure 21a it can be seen that at the highest altitude in the region of 60° to 65° N the Sr-90 background rose from about 150 to 225 dpm/1000 scf, so that by December over 30% of the amount present is from a recent influx. While part of this may have been from the equatorial region, the increase at lower latitudes is not as pronounced[5] and suggests that the equatorial region is not the major source of Sr-90 increase. Using the Ce-141/Sr-90 ratio it can be seen from Figure 35 that on 1 June 1960 the tropical average was 5.0 while the north polar average at the high altitudes was about 6.6 Assuming that Teak and Orange had an initial production ratio of 39 and is unfractionated, one would expect this debris to have a value of about 8.8 on 1 June 1960. Assuming further that the increase in concentration of cerium-141 was due only to influx from Teak and Orange, it appears that as an upper limit, 40% of the strontium-90 activity in the northern polar stratosphere between 60,000 and 70,000 feet was produced by Teak and Orange. While some of this cerium-141 may have come from debris from the Soviet test of 1958, other evidence suggests most of the Soviet debris was removed from the stratosphere by this time. Conclusive evidence provided by rhodium-102 data that debris from at least Orange and probably also Teak entered the lower stratosphere in the fall of 1959 will be presented later in this chapter.

Ashcan Data

The Ashcan data shown on Figure 35 indicates that sometime during the winter of 1958 - 1959 fresh debris entered the northern polar stratosphere at the 80,000 and 90,000 foot levels. The Ce-141 Sr-90 ratios at 80,000 feet rose to values similar to that associated with the HARDTACK-GRAPPLE debris while the ratios at 90,000 feet

rose to values more characteristic of Teak and Orange or Soviet debris. Salter[42] has indicated that starting in July 1959 the debris at 90,000 feet started mixing downward and that by October the Ce-144/Sr-90 ratio at 80,000 feet was the same as at 90,000 feet (See Figure 36). It was the month of October 1959 that marked the beginning of the rise in the Ce-144/Sr-90 ratio at 60,000 to 70,000 feet in the northern polar HASP measurements (Figure 35) so these Ashcan data are not inconsistent with the HASP data.

Soviet Debris vs Teak and Orange

While it cannot be categorically shown that the high values of the Ce-144/Sr-90 ratio at 90,000 feet in the spring of 1959 are definitely from the Soviet series alone or from Teak and Orange alone, downward mixing from Teak and Orange is strongly suggested and it is possible that these 90,000 foot Ashcan collections of March and April at Sioux City are the first recorded collections of debris from Teak and Orange. There are several additional reasons for believing that the high Ce-144/Sr-90 ratios in the Ashcan collections are not wholly from Soviet debris.

First, it seems unlikely that debris from the Soviet series would appear at all altitudes except 80,000 feet. Murayama[39] has shown rather convincingly that the large increase of strontium-90 at 80,000 feet in the northern stratosphere was almost exclusively from testing in the equatorial regions (See Figure 21b). The tungsten marked debris did not rise as high as did the average strontium-90 injections, consequently the material entering the 80,000 foot level had a lower than normal tungsten concentration. At 90,000 feet the Sr-90 increase was not as pronounced as that at 80,000 feet but surprisingly the debris at 90,000 feet seemed to have more than its share of tungsten. One possibility that suggests itself is that the Ashcan sampler had a lower collection efficiency for strontium-90 at the highest altitudes (Some reasons for this possibility

have been outlined in Chapter III). This notion is supported by the fact that, in the past, the strontium-90 profile has diminished at higher altitudes faster than has the carbon-11 profile measured at the same time[2]. In addition, Murayama[13] has also shown that Cs-137/Sr-90 ratios in Ashcan samples are higher than those in the HASP samples and also higher than the theoretically expected value (2.5 vs. 1.8), again suggesting lowered strontium-90 collection efficiency. In other words, the definite possibility exists that the high Ce-144/Sr-90 ratios observed at 90,000 feet are not representative of the debris sampled.

The second argument against a large quantity of Soviet debris remaining in the high northern stratosphere is the fact that a large fraction of the Soviet injection of 1958 can be accounted for in rainfall and lower stratospheric HASP samples through November 1959. Using a Sr-89/Sr-90 ratio of 160 at time of formation and the increased HASP strontium-90 inventories described above as modifications, the method of inventory used by Telegadas[11] suggests that 0.85 $\pm$ 0.15 megacuries of Sr-90 originating from the 1958 Soviet series can be accounted for below 70,000 feet. Probably no more than 0.25 megacuries of the original production from the Soviet series remains unaccounted for.

The final argument against there being large quantities of Soviet debris in the higher stratosphere as late as mid-1960 is the rather symmetrical distribution of strontium-90 between hemispheres. The highest concentrations are found at the highest altitudes and at the highest latitudes with the total inventory in the Northern Hemisphere being no more than 35% greater than that of the Southern Hemisphere. If any Soviet debris migrated into the Southern Hemisphere it must have done so at a level above 70,000 feet as there is little evidence at all of this debris below 70,000 feet in the equatorial region. In addition, the Ce-144/Sr-90 ratios during mid-1960 do not differ too markedly between the Northern and Southern Hemisphere.

Figure 37 shows a representative sampling of the Ce-144/Sr-90 ratios obtained during Phase V sampling. (See also Tables V and VI) While the Northern Hemisphere values are 10 to 20% higher than the Southern Hemisphere values at 40° latitude, the highest values are found at the highest latitudes and highest altitudes in both hemispheres. Further discussion on this point will be found in the section on rhodium-102 below. It is noted also that the balloon values again are somewhat higher than the aircraft values at equivalent altitudes.

While all of the above arguments by no means prove that no Soviet debris is left in the atmosphere they certainly tend to show that some other source of debris, namely Teak and Orange, is contributing to the concentrations found at high latitudes. It seems reasonable to assume that at least half of the increase in concentration of strontium-90 at the high latitudes during the winter of 1959-1960 was due to Teak and Orange. This assumption is supported by the results of measurements of rhodium-102 described below. If the assumption is correct the upper limit of the strontium-90 increase due to Teak and Orange of 10% as suggested earlier in this chapter is reduced to 20%.

Distribution of Rhodium-102

In the previous report (DASA 532), stratospheric concentrations of rhodium-102 were reported for the period July 1958 to August 1959. During that period the maximum concentrations were found in the tropical stratosphere along with concentrations in both the polar stratospheres one-half to one-quarter that of the tropical region. Since that time the picture has changed radically.

Apparently there have been two distinct sources of rhodium-102 injected into the stratosphere. Approximately 3 megacuries of rhodium-102 were produced in Orange, the second high yield rocket shot[38] fired at the end of the HARDTACK series.

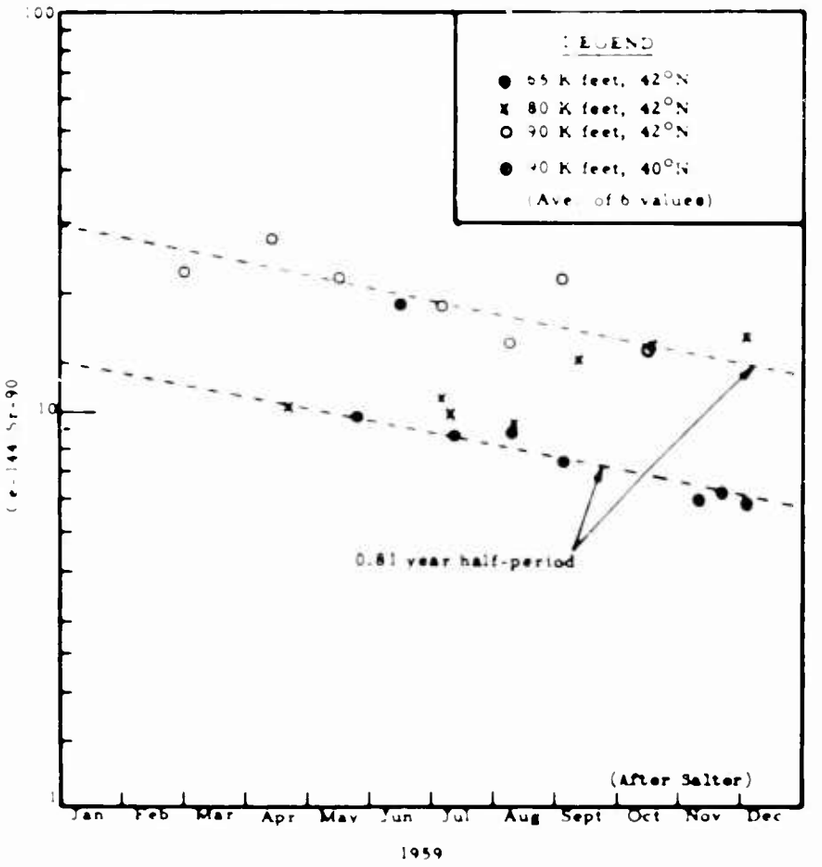
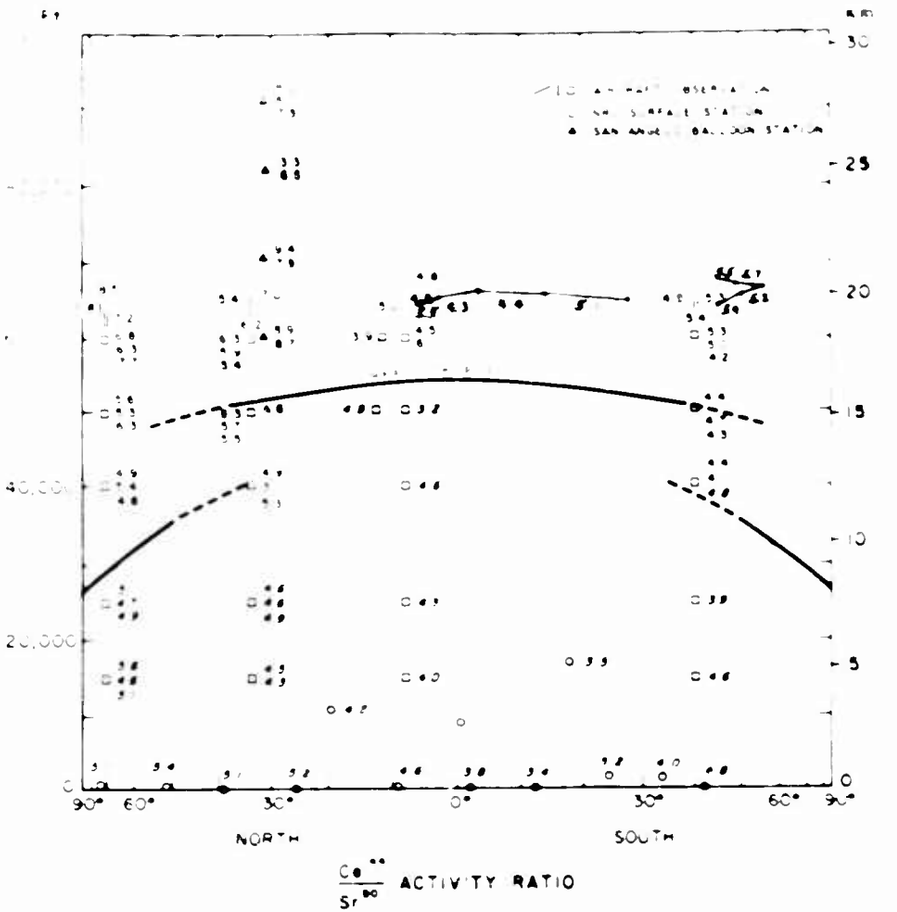


Figure 36 Ashcan Ce-144/Sr-90 in the Stratosphere at 42°N.



$\frac{Ce^{144}}{Sr^{90}}$ ACTIVITY RATIO

MAY 1960

FIGURE 37

It is estimated that as much as 0.3 megacuries were produced in several other low altitude bursts of the HARDTACK series. This latter source may have injected as much as 0.15 megacuries into the stratosphere with an ultimate distribution similar to that of tungsten. It is unlikely that more than 0.02 megacuries from this source remained in the stratosphere after May 1960.

According to data obtained by Kalkstein at AFCRL through the analysis of filter samples of stratospheric air, the stratospheric concentrations of rhodium-102 in the Northern Hemisphere increased during the autumn of 1959 to values above those attributable to injections of rhodium-102 by HARDTACK surface bursts. Measurements of stratospheric air in the Southern Hemisphere by Kalkstein* suggested that a similar increase may have occurred there in the autumn (May, June) or winter (July, August) of 1959 and that by the autumn (June)of 1960 the rhodium-102 concentrations in the Southern Hemisphere equalled those in the Northern Hemisphere.

A number of HASP samples collected during the winter of 1959-1960 in the Northern Hemisphere have been analyzed for rhodium-102. The mean concentrations indicated by these samples, arranged according to the 5000 foot altitude layer and according to the region (south of Ramey, south of Laughlin, between Laughlin and Minot, or north of Minot) in which they were collected, are given in Figure 38a. It is evident that the highest concentrations occur at the highest latitudes and highest altitudes. This latitudinal distribution of activities is confirmed by the AFCRL data.

It appears that the rhodium-102 is being brought down from its original injection site by strong vertical mixing in the polar stratosphere during the "winter" season (late autumn to early spring). The vertical concentration gradient suggests that vertical mixing rather than subsidence of air is the main factor in the

*Paper presented at [illegible] per Atmospheric [illegible] Albuquerque (April 1961) AFCRL-420

appearance of rhodium-102 below 70,000 feet. The rhodium-102 in the tropical stratosphere probably reached there through downward mixing into the polar stratosphere and then lateral mixing into the tropical stratosphere. The total amount of rhodium-102 in the stratosphere of the Northern Hemisphere during the winter of 1959-1960 was about 0.25 megacurie (corrected for decay to August 1958). Thus, during the second year after its injection at high altitude about 9 percent of the rhodium-102 activity was carried down into the lower northern stratosphere. If the assumption is made that an equal amount of rhodium-102 resided in the lower Southern Hemisphere by May 1960, then 0.52 megacurie of rhodium-102 or 18% of the activity from Orange (and probably from Teak) was below 70,000 feet. Figure 38b shows the rhodium-102 measurements made during May 1960. List[38] has calculated that this distribution represents a total stratospheric inventory between 10,000 and 70,000 feet of 0.37 megacuries. This compares favorably with the estimate made above when consideration is made of possible amounts that have fallen below 10,000 feet since September 1959 (when rhodium-102 was detected in ground level air near Chicago by Gustafson[40]). In addition, it is seen in Figure 38b that the rhodium-102 is distributed symmetrically about the equator in the range that the sampling has been conducted. There is a possibility that this "snapshot" of the stratosphere is not representative of the whole year and that seasonal effects will produce differences not apparent here.

An initial estimate of the residence half-time of material placed in the mesosphere is afforded by the rhodium-102 data. If the assumption is made that a few months (for instance, 3 months) are required for the debris to be mixed to any extent through the mesosphere then it is seen from the above information that about one-sixth of the rhodium-102 from Orange departed from the mesophere within 18 months after mixing. This represents a residence half-time of 68 months or about 5½ years

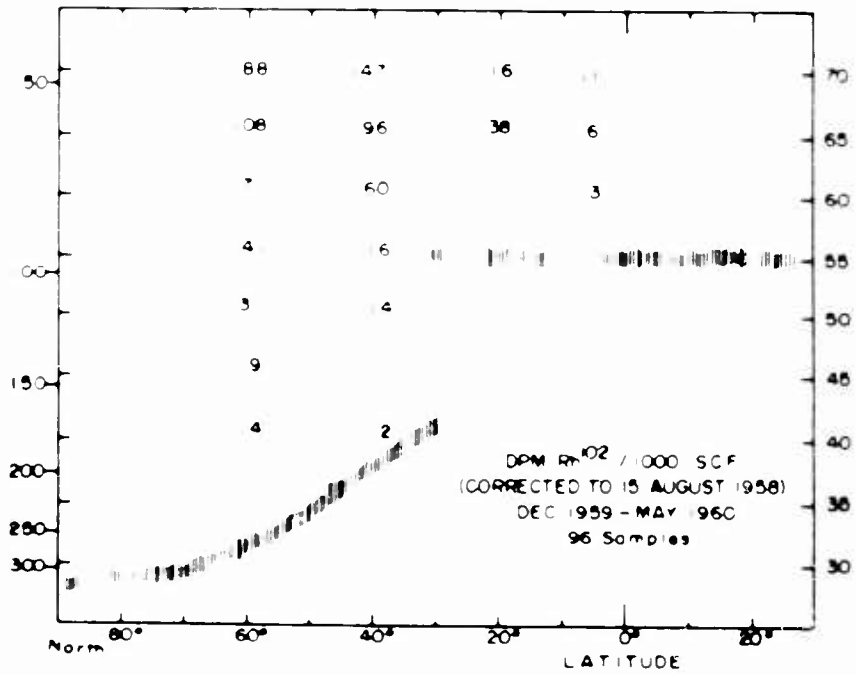


Figure 38a MEAN DISTRIBUTION OF RHODIUM - 102, DECEMBER 1959 - MAY 1960

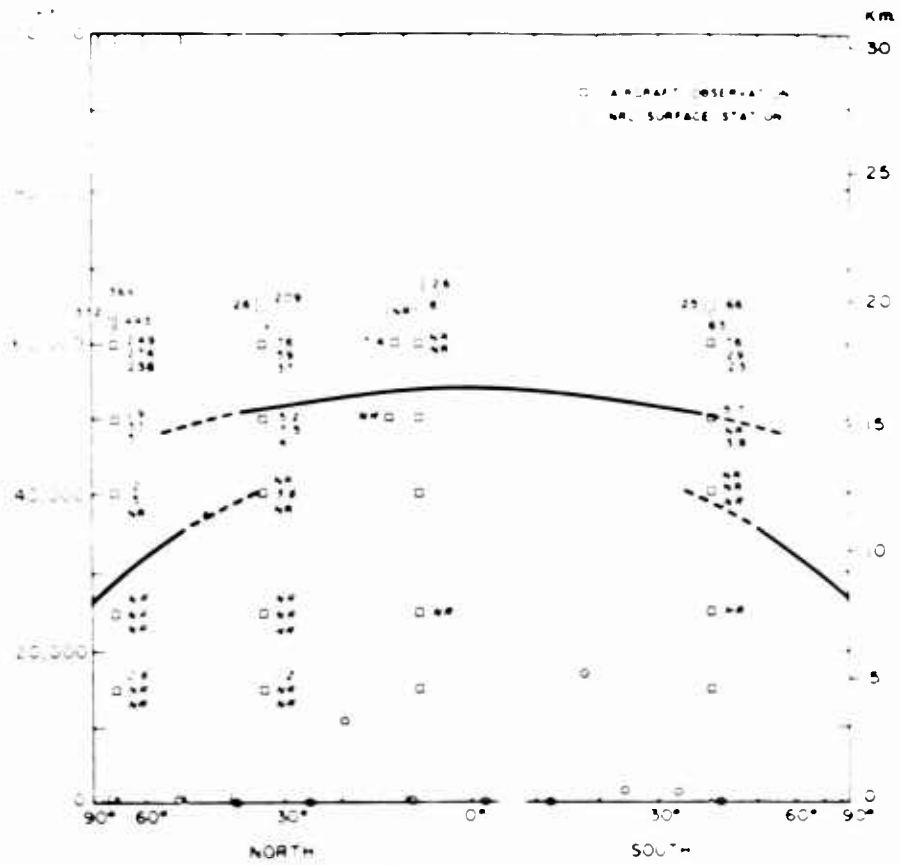


Figure 38b Rh^{106} d/m/1000 sc. corrected for decay to Aug 12, 1958) MAY 1960

(mean residence time of 8 years).

The rhodium-102 data also affords an opportunity to evaluate in a more quantitative manner the source of strontium-90 and cerium-144 concentrations found in the polar stratosphere. If the assumption is made that Teak and Orange debris contains the major fraction of the 0.4 megacuries of strontium-90 originally placed in the mesosphere[28], then for every 300 dpm of rhodium-102 on 12 August 1958 there would be on 1 June 1960, 38 dpm of strontium-90 and, assuming the initial production of cerium-144 shown in Table VIII, there would be on 1 June 1960, 318 dpm of cerium-144. When the Teak and Orange debris is subtracted from the values shown in the May 1960 "snapshot" (Table VI and Figure 37) by using decrements proportional to the rhodium-102 values, the Ce-144/Sr-90 ratios that remain average about 4.7 in the Southern Hemispheric and northern equatorial stratosphere and average about 5.7 in the northern polar stratosphere. The decrements amount to about 25% of the strontium-90 at 70° North, 10% at 10° North and 40° South, and about 2% at the equator. These figures are comparable to the 20% to 10% figures previously suggested for the amount of strontium-90 in the northern stratosphere from Teak and Orange deduced from strontium-90 and Ce-144 data. The Ce-144/Sr-90 ratios of the remainder imply that the debris in the northern polar stratosphere exclusive of Teak and Orange is about 80 days younger than that found elsewhere in the stratosphere. This result suggests that some of the debris in the northern polar stratosphere came from Soviet tests, although it is not altogether conclusive. For instance, if another decrement similar to that assigned Teak and Orange is applied to the previous remainder to "remove" the Soviet debris, the strontium-90 concentrations in the Northern Hemisphere fall to values about 30% lower than those in the Southern Hemisphere. This is an unlikely situation although it could actually prevail if the Southern Hemisphere were receiving a large austral

winter influx of older material from the high equatorial region. On the other hand, even before the late 1958 Soviet series, HASP measurements have shown that debris in the Southern Hemisphere has always tended to be older than in the Northern Hemisphere.

While the rhodium-102 data can yield valuable information on the rates and location of entry into the lower atmosphere of debris from Orange, there are several uncertainties in the data. First, the actual amount of production is not known with certainty. Fortunately, rhodium-102 is not a fission product since it is shielded in the decay of mass chain 102 by stable ruthenium-102 (rhodium-102 is produced by the n, 2n reaction in natural rhodium-103). Second, the decay scheme of rhodium-102 is not well known. Rhodium-102 decays by K and L capture as well as by negative beta emission. An additional complicating factor is the presence of rhodium-101 and a long lived isomer of rhodium-102 which has an ill-defined half life of several years and makes a correction for decay to the anniversary date uncertain. The empirical decay curve obtained by Kalkstein at AFCRL has been used to correct rhodium counting data on HASP samples for their rhodium-102 concentrations and these have been extrapolated back to 12 August 1958 using a 210 day half life. Finally, concentrations of rhodium-102 have been somewhat low and long exposures of the filter paper are required to obtain good counting statistics. Often several papers collected over a relatively large range of latitudes are pooled to increase the amounts of rhodium-102 available for measurement.

Distribution of Plutonium

Plutonium has been measured in a number of HASP samples collected up to mid-1959. The plutonium samples were measured in separate aliquots of the original sample solution of the filter paper. After selective purification, the plutonium is electrodeposited carrier-free on stainless steel disks (See Figure 39). Radiometric assay

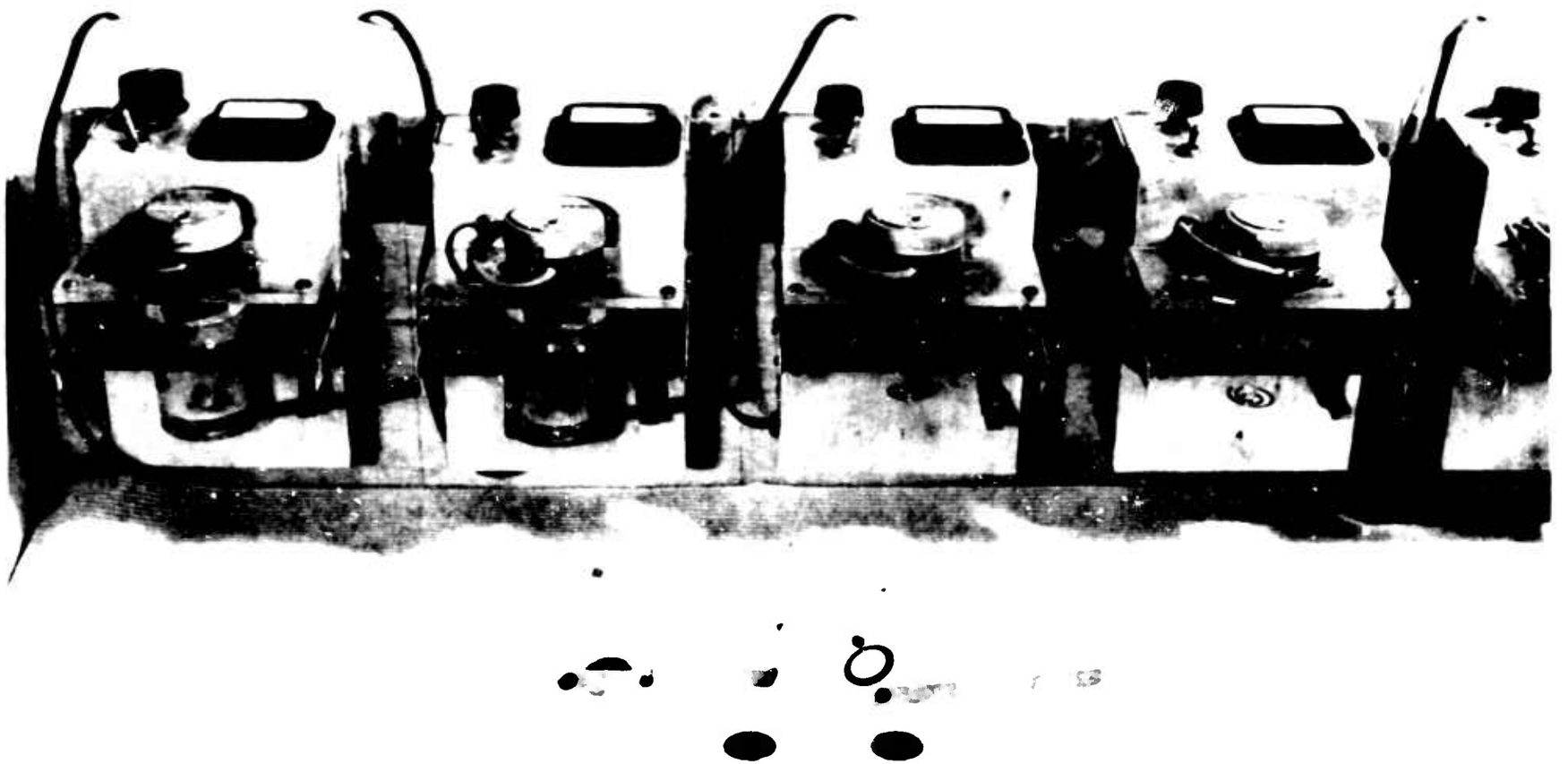


FIGURE 39 ELECTRODEPOSITION APPARATUS

is performed by scintillation counting of the alpha emissions. Since the alpha spectrum is not measured, no discrimination between plutonium-239 and plutonium-240 is made.

Figure 40 shows the distribution of the Pu/Sr-90 ratios in 228 HASP samples collected over a period of 18 months. The mean value is 0.018 dpm Pu per dpm Sr-90. Variations from this mean are larger and more abundant than was true for the Cs-137/Sr-90 ratios reported in DASA-532. Real variations in the Pu/Sr-90 ratios are not unexpected since this ratio for debris depends upon the capture/fission ratio in the weapon which produced it. Even so the extreme values shown in the Figure are probably the result of analytical error.

In Table XI the Pu/Sr-90 ratio is tabulated according to the origin or sampling date of the debris. Certain tests, especially the fall 1958 UK test, produced debris with a low Pu/Sr-90 ratio. Debris in the Southern Hemisphere had a higher Pu/Sr-90 ratio than debris in the Northern Hemisphere during late 1958 and early 1959. This probably resulted from a greater representation in the Southern Hemisphere of background debris from high yield weapons, especially from the CASTLE tests which would have a high capture/fission ratio. This substantiates the conclusion reached above in the discussion of the strontium-90 distribution that debris sampled in the southern polar stratosphere especially was originally injected into the high tropical stratosphere.

FRAMES

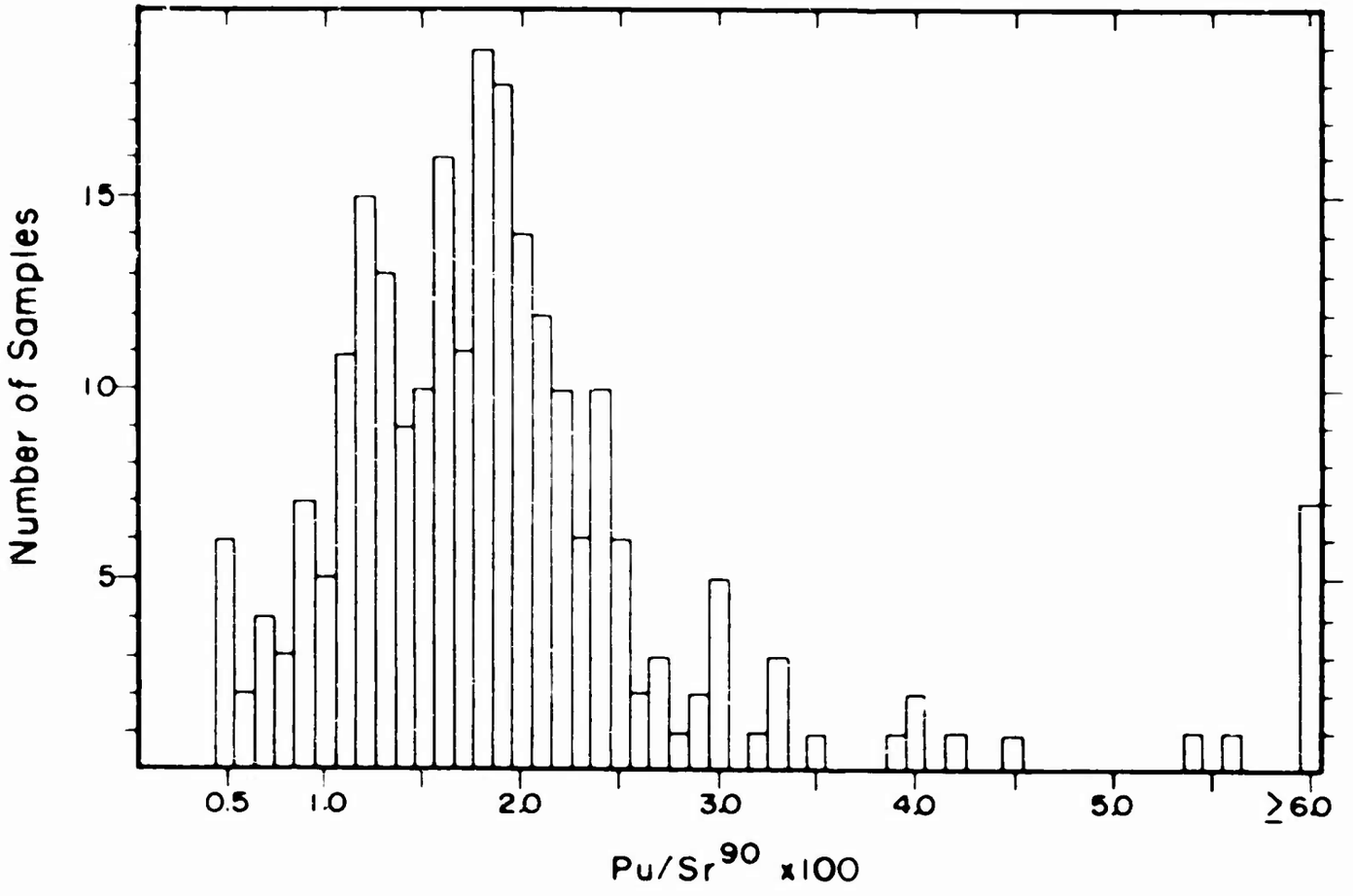


FIGURE 40 The Distribution of Pu/Sr^{90} Ratios in HASP Samples

TABLE XI

Variations in the Plutonium/Strontium-90 Ratio of Stratospheric Debris

	No. of Samples	100 Pu/Sr^{90}
Northern Polar Stratosphere		
Background debris, November 1957	4	1.6 ± 0.3
Debris from fall 1957 Soviet tests	3	2.1 ± 0.3
Background debris, March-April 1958	12	1.8 ± 0.3
Debris from spring 1958 Soviet tests	3	1.4 ± 0.5
Background debris, May 1958	9	2.1 ± 0.4
Background debris, September-October 1958	12	1.9 ± 0.9
Debris from fall Soviet tests	6	1.2 ± 0.5
Background debris, January-February 1959	2	1.5 ± 0.1
Background debris, March-April 1959	26	1.6 ± 0.4
Background debris, May 1959	3	1.9 ± 0.1
Tropical Stratosphere		
Background debris, November 1957	2	1.2 ± 0.1
Debris from fall 1957 British tests:		
1. Intercepted during November 1957	4	1.4 ± 0.3
2. Intercepted during February 1958	4	1.2 ± 0.2
Background debris, March 1958	4	1.7 ± 0.2
Debris from spring 1958 Soviet tests	3	1.6 ± 0.4
Background debris, May-June 1958	8	2.3 ± 0.2
Debris from HARDTACK:		
1. Intercepted during June 1958	1	3.0
2. Intercepted during September 1958	4	1.6 ±0.9
Debris from fall 1958 British tests	10	0.7 ±0.2
Background debris, 30° North to 15° South latitude:		
November-December 1958	9	1.3 ±0.3
January-February 1959	12	1.5 ±0.5
March-April 1959	8	1.6 ±0.6
May 1959	2	1.8
Background debris, 15° South to 30° South latitude:		
November-December 1958	4	1.9 ±0.5
January-February 1959	2	3.7 ±2.4
March-April 1959	4	2.8 ±1.3
May 1959	1	2.2
Southern Polar Stratosphere		
Background debris, September-October 1958	4	2.3 ±1.0
Background debris, November-December 1958	6	2.6 ±0.7
Background debris, January-February 1959	9	1.8 ±0.3
Background debris, March-April 1959	6	2.8 ±0.8
Background debris, May 1959	2	3.7 ±0.5

CHAPTER VII

NATURAL RADIOACTIVITY IN THE ATMOSPHERE

Introduction

With the drop in rate of weapons testing since 1958, naturally radioactive materials either produced in the stratosphere by the action of cosmic rays or exhaled from the surface of the earth may become increasingly important as trace materials for the study of the behavior of the atmosphere. In addition, the source of the natural radioactivity is more evenly distributed around the world than fission products and the materials have had sufficient time to establish equilibrium distributions, if such exist. Measurements of the gradients of these distributions can cast considerable light on the rate and direction of various possible mixing processes. Unfortunately, the natural background of several isotopes such as carbon-14 and tritium has been overwhelmed by the production of these materials in weapons detonations. An attempt will be made in this chapter to outline the information that has been gained in studying the distribution of carbon-14, tritium, beryllium-7, phosphorus-32, and radon daughter products such as lead-210.

Carbon-14

Carbon-14 is produced in the stratosphere by the interaction of cosmic ray neutrons with nitrogen in the air. While the reactions probably take place mainly between 50,000 and 100,000 feet, the 5760 year half life of carbon-14 is so long compared to mixing rates in the atmosphere that a natural gradient of this material is not expected. Since the stratosphere contains about 311 ppm of carbon dioxide and the normal stratospheric carbon-14 content is about 9×10^{27} carbon-14 atoms[18], the natural concentration should be about 71×10^{5} atoms of C-14 per gram of air or 13 dpm per gram of carbon.

When neutrons escape from a weapon into the air, carbon-14 is formed in a manner similar to the natural processes. Libby[46] has estimated that the stratospheric burden of carbon-14 had doubled as a result of weapons testing up to 1 January 1958. This was borne out by balloon measurements reported by Hagemann[45] who also estimated the total bomb production of carbon-14 by the end of 1958 to be 25×10^{27} atoms.

The stratospheric distribution of carbon-14 reported by Hagemann was compared with the HASP and Ashcan results in the previous report (DASA-532). Briefly, it was noted in that report that the excess carbon-14 profiles showed a maximum at 80,000 to 90,000 feet at most of the sampling sites as is suggested by the strontium-90 data. The size of the peak at this altitude was greater in the carbon-14 profiles than in the strontium-90 profiles. This may be due to a possible decrease in sampling efficiency for strontium-90 in the Ashcan at higher altitudes which is not being taken into account. If the HASP data are extrapolated upward using the carbon-14 profile rather than the Ashcan profile, the HASP stratospheric inventories would be increased by perhaps 25%. The stratospheric mean residence time for debris injected into the high equatorial region based on the carbon-14 data is about 4 years. This is not vastly different from the value based on measurements of particulate debris. The carbon-14 data indicate that, prior to the October 1958 injections, the Soviet injections were all stabilized at low altitudes in the polar stratosphere and they made no long range contribution to the stratospheric burden of nuclear debris. Finally, it was noted that the carbon-14 exhibited a poleward transfer of debris from the equatorial region preferentially during the winter season in each hemisphere.

Since January 1960 a number of samples of carbon-11 in CO_2 taken from surface air have been measured as part of the HASP program. The CO_2 is purified in a vacuum system (Figure 11). Oxygen and nitrogen are removed by pumping from the frozen CO_2. After reaction with Ca O, radon is removed by further pumping. The Ca CO_3 is decomposed by heating and the CO_2 is analyzed by proportional counting at two atmospheres in a well shielded anticoincidence counting apparatus (Figure 12).

The results of the measurements are shown in Table XII and lie between the 15% and 30% values predicted by Libby and Hagemann. Surface air during early 1960 was enriched in carbon-11 by about 20% over the concentration which would have existed had there been no production of that radionuclide by nuclear weapons testing. This is in agreement with data obtained by Munnich at Heidelberg where the excess carbon-11 reached +30% during mid-1959 declining to about +20% by the end of 1959. It also agrees with data obtained by Broecker for three sites in the Northern Hemisphere during late 1959 and early 1960. Concentrations in the Southern Hemisphere may still be a few per cent lower than those in the Northern Hemisphere according to data obtained by both Munnich and Broecker. There is only a slight suggestion of a spring peak during March and April 1960. The spring peak in tritium described below was much more pronounced. A possible peak in carbon-11 may have been partially obscured by a wintertime increase in the use of fossil fuels depleted in natural carbon-11 (Suess effect). A drop in concentration is noted starting in September. This suggests that the rate of removal from the stratosphere is diminishing during the fall which has been the case for particulate debris and tritium. By early 1961, the concentrations fell to around 10% but then returned to the 20% level by February. This drop may be due to a combination of winter Suess effect and mixing into the ocean. It is apparent that the concentration in the troposphere has been diminishing somewhat faster than had been

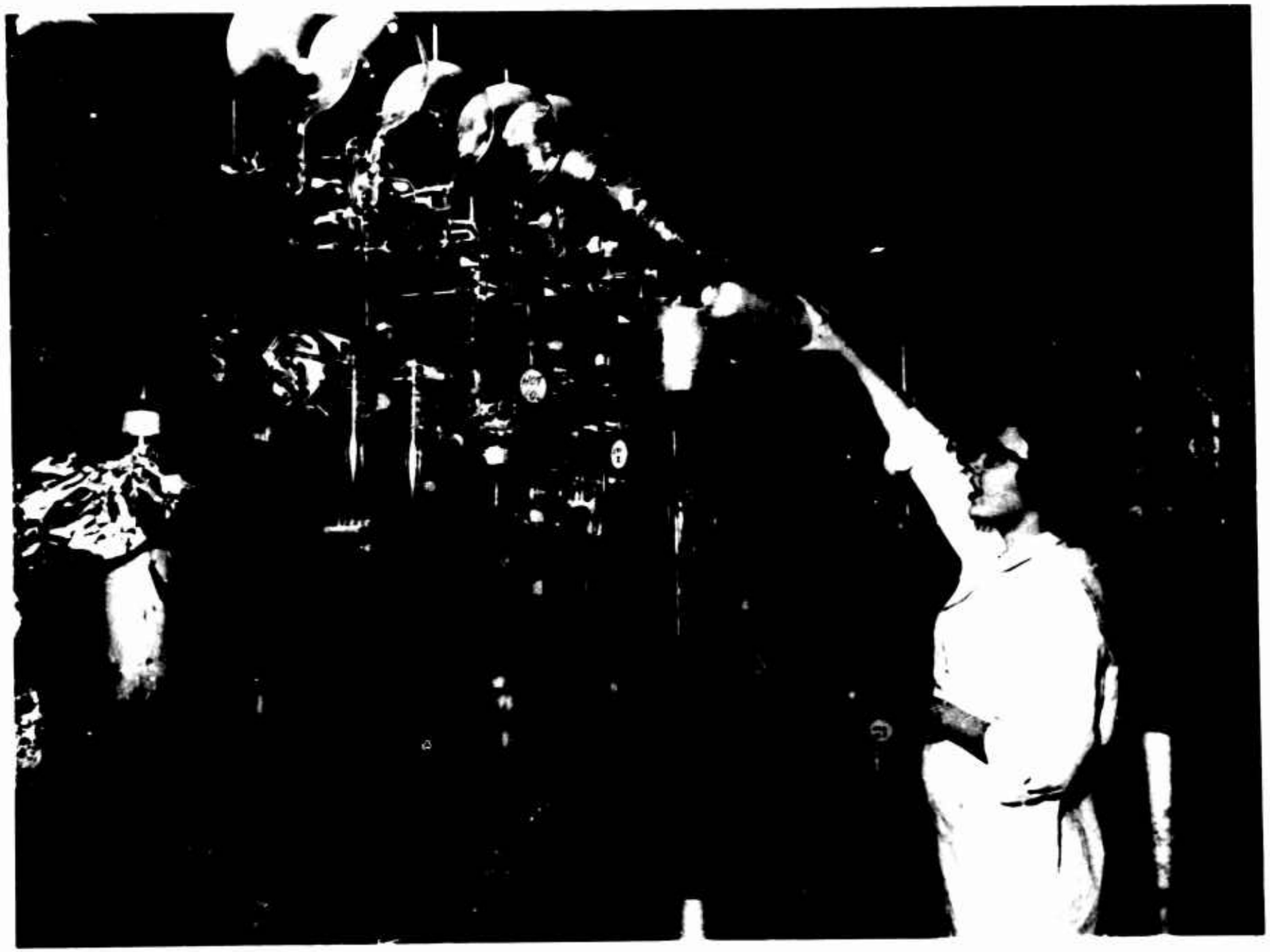


FIGURE 41 VACUUM PURIFICATION SYSTEM

FIGURE 42 ANTI-COINCIDENCE COUNTING APPARATUS

Table XII

Radiocarbon Concentrations in Tropospheric Air at Washington Township, New Jersey

Collection Period	ΔC^{14}*
19 Jan - 25 Jan 1960	+20.85%**
2 Feb - 9 Feb 1960	+20.00%
23 Feb - 1 March 1960	+18.45%
1 March - 8 March 1960	+22.43%
29 March - 5 April 1960	+22.40%
5 April - 12 April 1960	+16.00%
26 April - 3 May 1960	+18.24%
24 May - 31 May 1960	+20.82%
28 May 1960 (oak leaves)	+17.78%
21 June - 26 June 1960	+19.73%
26 July - 2 Aug 1960	+20.76%
23 Aug - 6 Sept 1960	+18.10%
6 Sept - 20 Sept 1960	+19.57%
27 Sept - 4 Oct 1960	+17.19%
4 Oct - 11 Oct 1960	+17.19%
1 Nov - 17 Nov 1960	+14.09%
1 Dec - 11 Dec 1960	+12.02%
31 Dec 1960 - 18 Jan 1961	+ 9.04%
18 Jan - 25 Jan 1961	+15.42%
18 Jan - 25 Jan 1961 (Closter, N. J.)	+14.40%
1 Feb - 8 Feb 1961	+19.13%
16 March - 3 Apr 1961	+20.42%
16 Apr - 1 May 1961	+15.63%

* Excess of Standard = 0.95 x NBS oxalic acid

** Counting error = $\pm$ 1.5% except for samples collected 5-12 April 1960, and 18 Jan - 25 Jan 1961 (Closter, N. J.) which, because of small size, had an error of $\pm$ 2.5%

previously expected.

Tritium

Natural tritium in the atmosphere results from the spallation of atmospheric nuclides by energetic cosmic ray particles. Recent satellite measurements indicate that tritium may also be a component of solar flares. The half life of tritium is relatively short (~12.4 years); consequently, there may be a noticeable gradient of natural tritium in the stratosphere. Tritium is also introduced into the atmosphere by the explosion of thermonuclear weapons. Some tritium may be present initially in the weapon and some tritium may be produced during the course of the thermonuclear reactions.

Only a limited number of tritium measurements have been made in the stratosphere. Those reported by Hagemann[45] indicated that tritium had a positive linear correlation with excess carbon-14 atoms such that the T/C-14 atom ratio was 0.4. The deduction was made that on 1 January 1958 the stratospheric inventory was 2.4×10^{27} bomb produced tritium atoms and 0.6×10^{27} natural tritium atoms. This latter figure amounts to 7×10^{5} tritium atoms per gram of air and leads to a natural production rate of 1 tritium atom per square centimeter per second. Assuming a water vapor concentration throughout the stratosphere with a frost point of about -75° C, the natural stratospheric concentration of tritium in water would be about 10^{6} T.U. (tritium units) or 10^{6} tritium atoms per 10^{18} hydrogen atoms. This number can be compared with prebomb continental rains which contained about 9 T.U. and the oceanic tritium content which is about 1 T.U.[47].

Since 1 January 1958 there have been several sources of injection of tritium into the stratosphere. Libby[46,48] has suggested that little tritium is injected into the stratosphere from surface bursts in the tropics (especially from water

surface bursts). The reason for this is the fact that the especially cold tropic tropopause acts as a cold trap and prevents the entry of most of the water (including T_2O or HTO). Indeed, the tritium concentration in rains falling after the 1954 CASTLE test series died away with a 35 day half life suggesting a tropospheric source for this material. However, as Libby points out[46], Teak and Orange debris was not subjected to the tropopause scavenging action and consequently may have raised the natural level of stratospheric tritium several hundred fold. In addition, the Soviet shots of October 1958 having been fired in cold dry polar air may have left more in the stratosphere than other tropospheric explosions. Libby[46] predicted that spring and summer rains in 1959 could reach a value of 1000 T.U.

Measurements of tritium in rainfall have been made as part of the HASP program since April 1959. After collection in receivers similar to those shown in Figure 43, the rain samples are distilled and then enriched electrolytically. Analysis is performed in the proportional counting apparatus shown in Figure 42. The results are tabulated in Table XIII and are plotted in Figure 44. As predicted by Libby, there has been a spring rise in 1959 with concentrations reaching at least 1400 T.U. A similar rise was noted in Chicago rain in the spring of 1956 after the 1955 Soviet test but not in the spring of 1955 after the 1954 CASTLE test series. During the summer and fall of 1959 the tritium concentration diminished exponentially with a half life of about 40 days. This suggests that little tritium was entering the tropopause during this period. Starting in December 1959 the concentration of tritium in rain started to rise exponentially with a doubling time of about 55 days until a peak was reached in July 1960 and then the characteristic 40 day depletion rate was again established.

FIGURE 43 RAINFALL COLLECTION RECEIVERS

Table XIII

Tritium Concentrations in Precipitation at Westwood, New Jersey

Collection Date	Inches of Rain	Tritium Units ($T/H \times 10^{18}$)
28 Apr 59	0.95	1435
1 May 59	0.34	1370
13 Jun 59	1.00	843
14 Jul 59	0.22	650
9 Aug 59	2.10	148
2 Sep 59	0.92	38.5
24 Oct 59	1.41	86.5
25 Nov 59	1.33	46.5
29 Dec 59	1.10	100
14 Jan 60	0.82	42.0
11 Feb 60*	1.02	62.9
11 Feb 60*		65.6
26 Feb 60	1.07	102
18 Mar 60	0.28	76.0
31 Mar 60	0.74	123
27 Apr 60	0.40	193
12 May 60	0.28	213
31 May 60	0.30	282
18 Jun 60	0.20	249
2 Jul 60	0.48	432
15 Jul 60	0.84	431
31 Jul 60	2.80	190
11 Aug 60	1.49	249
13 Sep 60	5.01	100
17 Oct 60	0.50	75

Analytical error is about $\pm$ 10%

* Duplicate analysis

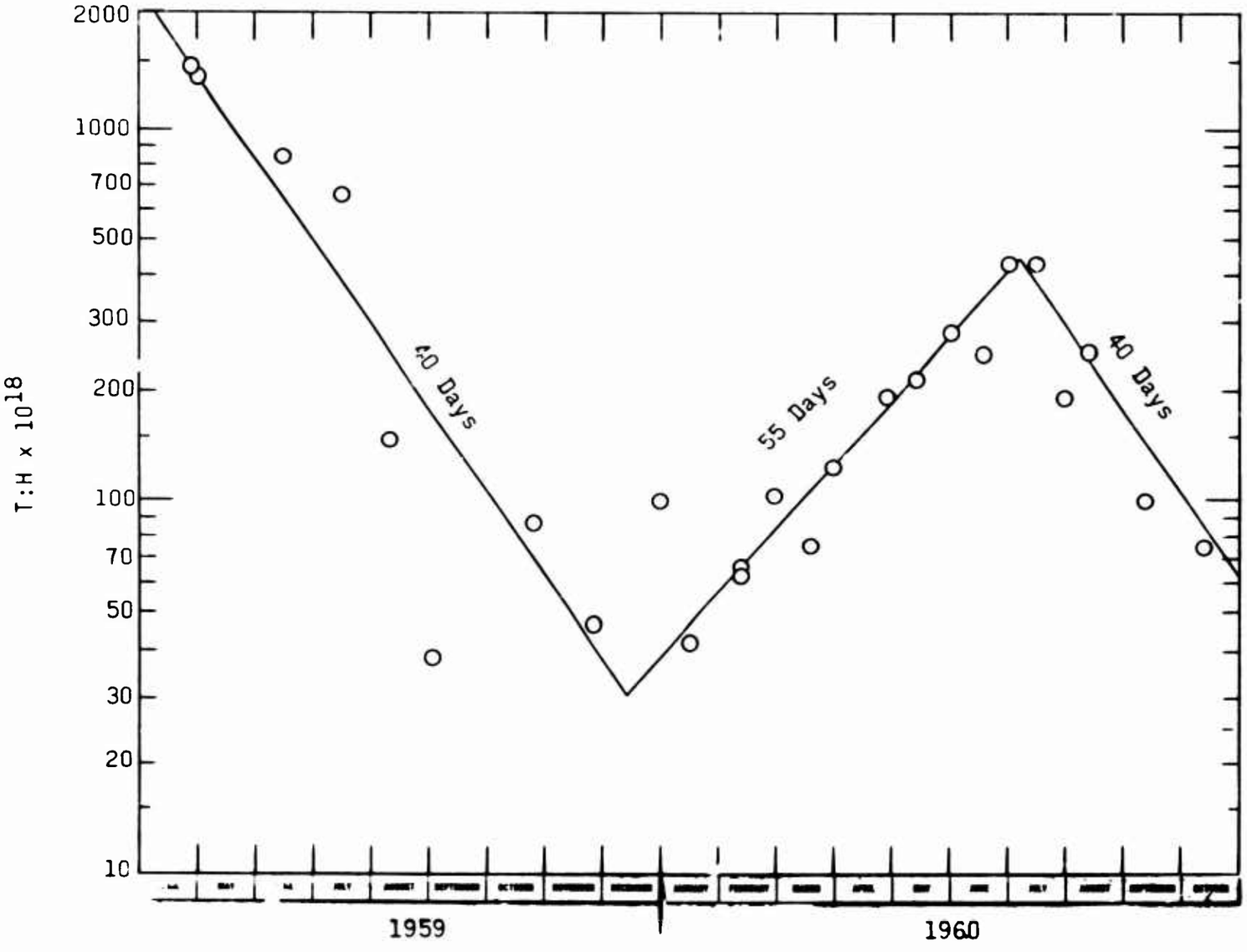


Figure 44 TRITIUM CONCENTRATIONS IN RAINFALL AT WESTWOOD, BERGEN CO., NEW JERSEY

Coupled with other evidence that debris from Teak and Orange had reached the lower stratosphere by the winter of 1959-1960, it is suggested that a small portion of this spring rise is from these high altitude rocket shots. If Libby's injection estimate is correct, the spring of 1961 and 1962 may show even greater increases of tritium in rain than in 1959 and 1960. An increasing trend in mid-stratospheric tritium concentration is borne out by balloon measurements between 80,000 and 100,000 feet over England made by F. Brown[19]. His results indicate a gradual rise in tritium concentration from about 4.5×10^6 atoms per gram of air at mid-1958 to about 7.2×10^6 atoms per gram of air at mid-1960. This is consistent with the picture of an influx from Teak and Orange.

An interesting side light of Brown's work is the persistent high water vapor concentrations noted at 90,000 feet. He measures a mixing ratio of about 0.04 grams of water per kilogram of air (F.P. = -76°C) while frost-point measurements at 50,000 feet over England have given mixing ratios of about 0.002 g/Kg (F.P. = -83°C). This distribution can be produced by strong vertical mixing in the polar regions which takes water vapor originally in the upper troposphere to the high latitude mid-stratosphere where it can mix equatorward. The low readings at 50,000 feet may be partially accounted for by the "sponging" action of the hygroscopic sulfate dust layer found above the tropopause (Chapter III) as well as by the action of a "cold trap" such as that found at the tropical tropopause. Since the sulfate concentration is about 4×10^{-6} grams per kilogram of air at 65,000 feet, the water/sulfate ratio is about 1200 grams per gram (M.R. = 0.005, F.P. = -82°C).* A completely satisfactory explanation of the observed stratospheric water vapor concentrations has not as yet appeared.**

*cf. Mastenbrook, H. J. and J. E. Dinger, Journ. Geophys. Res. 66, 1437 (1961)
**but see: Goldsmith, P. and F. Brown Nature 191, 1033 (1961)
and deTurville, C. M. Nature 190, 191, 156, 1183 (1961)
and especially Gutnick, M. Journ of Geophys. Res. 66, 2867 (1961)

One weakness must be mentioned concerning the HASP surface air and water collections and that is, of course, that they are point measurements in space. While it may be argued that a particular point on the earth may be representative of a large area, especially long after large scale testing has ceased, extreme caution must be exercised in making this generality. Considerable meteorological variability can exist from point to point in such items as rainfall and previous history of passing air masses. These factors can be taken into account properly only when comparisons are made with a number of other stations. In the case of tritium in rainfall, several additional complicating factors may exist. Machta[50] has pointed out that part of the Westwood rise may be due to increased amounts of tritium input from surface sources such as escape gases from the Savannah River plant coupled with seasonal flow from the south. Another source of the spring rise has been pointed out by R. M. Brown[51] and that is the increased evaporation of surface water during the spring and early summer. Brown has shown tritium concentrations in rainfall during 1959 and 1960 at Ottawa which are similar to those at Westwood. The levels of tritium in surface water have not shown the same large fluctuations, however. By mid-1959 the surface water had a tritium level of about 300 T.U. which gradually dropped to 250 T. U. by late 1960. It is seen that the surface concentration was higher than the rain concentration for a good deal of this period and since Brown estimates that as much as 10% of the surface reservoir may evaporate each year; this source can make a substantial contribution to the tritium in rain. This recycling mechanism apparently does not apply to particulate fission products, however. (See Chapter X).

Beryllium-7

Beryllium-7 is a spall product of high energy cosmic rays acting upon the the elements of the atmosphere. It apparently has not been produced artificially

in any noticeable quantity as a result of nuclear explosions[52,53]. Lal has predicted the equilibrium distribution of this material throughout the atmosphere as a function of latitude and altitude assuming a perfectly quiet atmosphere (See Figure 47). Since there is a marked production gradient in the atmosphere and since the half life of beryllium-7 (53 days) is relatively short, departures of the measured beryllium-7 concentrations from the estimated production concentrations should shed some light on the mixing processes in the atmosphere.

Measurements of Be-7 in surface air and rainwater have been made by a number of investigators[53-56]. Gustafson[40]* has made an extensive set of measurements of beryllium-7 and other nuclides in surface air filters collected at the Argonne National Laboratory near Chicago. The results of his beryllium-7 and cesium-137 measurements are shown in Figure 45. The most evident feature of these data is the pronounced spring rise in both 1959 and 1960 similar to that observed in the Westwood tritium data (Figure 44). Gustafson attributes the rise in cesium-137 to the disruption of the tropopause in the mid-latitude regions during the spring months resulting in an influx of high altitude debris, much of it of polar origin, into the lower troposphere. In addition, the rising tropopause in the spring encompasses more stratospheric debris. During the summer and fall the concentration of cesium-137 diminishes more or less exponentially showing tropospheric clearing of the large spring incursion. The minima in cesium-137 concentrations are considered to be a measure of the steady-state stratospheric drip upon which the springtime gush is superimposed. If this is the case, it is noted that the apparent stratospheric residence time is increasing. This situation is to be expected as debris originally injected at high altitudes in the stratosphere begins to predominate over that injected only a short distance above the tropopause.

*See also Gustafson, P. F., M. A. Kerrigan, and S. S. Brar, Nature 191, 454 (1961)

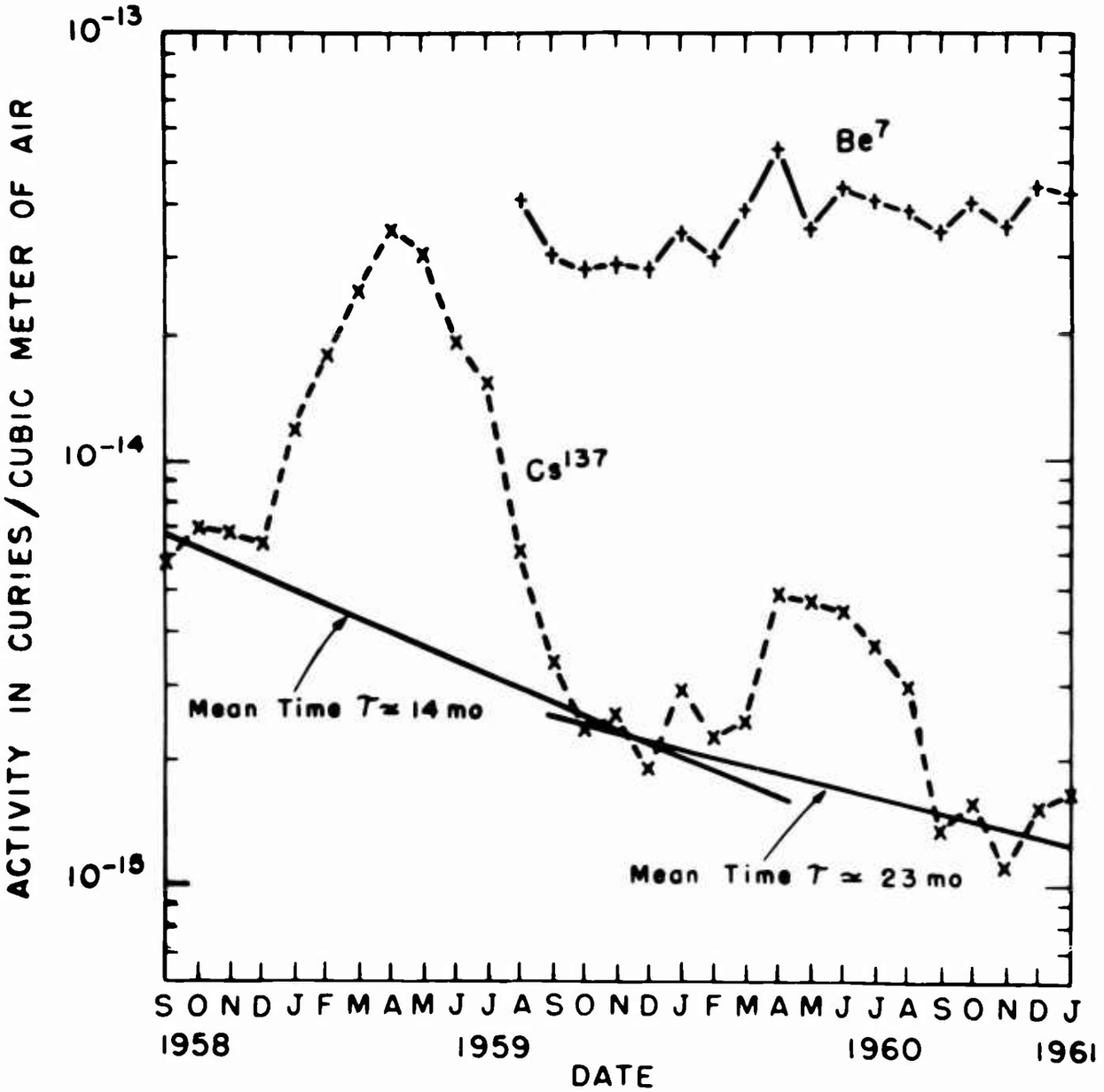


FIGURE 45 BE-7 AND CS-137 CONCENTRATIONS IN CHICAGO AIR

The beryllium-7 exhibits a spring rise as does the cesium-137 but it is not as pronounced. The reason for this is the fact that beryllium-7 is continually being produced in the troposphere (as well as in the stratosphere) while cesium-137 is not. Using the simplifying assumption that the concentrations noted in the air at Chicago are of "tropospheric" origin and represent the equilibrium concentration minus that scavenged by precipitation, Gustafson "added back" the portion removed by the rainfall and determined the mean effective equilibrium tropospheric concentrations in the summer and winter seasons respectively to be 25 and 14 beryllium-7 atoms per liter (7.7 and 4.3 dpm/1000 scf).* This difference is explained as indicating the increased size of the summer troposphere which includes the higher producing layer between 30,000 and 45,000 feet. It should be pointed out that this somewhat simplified picture makes no attempt to describe the vertical or latitudinal distribution witnin the troposphere but rather applies only to the eventual concentration in surface air. Limited data from Canada[56] and the U.K.[55] indicate concentrations of beryllium-7 in surface air which are not inconsistent with those measured by Gustafson. A number of tropospheric air samples of beryllium-7 were made using B-57 aircraft between 15,000 and 50,000 feet during March and April 1960[37]. When these data are fully analysed and correlated with other meteorological data they should shed considerable light on the springtime tropospheric distribution of this material.

A number of HASP samples collected during the winter and spring of 1959-1960 have been analysed for beryllium-7. Standard chemical separation and gamma ray spectroscopy techniques were used to determine the concentrations. The mean distribution is pictured in Figure 46. Lal's calculated equilibrium distribution of

* For Be-7 in air, 1 dpm/1000 scf = 2.7 atoms/gm = 3.2 atoms/liter = 1.35×10^{-14} curies/m^3 = 1.35×10^{-17} curies/liter.

stratospheric beryllium-7 concentrations, assuming a stagnant atmosphere, is shown in Figure 47 for comparison. The measured concentrations have roughly the same distribution as the equilibrium concentrations, but the activity levels are different. In the polar stratosphere and in the southern tropical stratosphere the measured concentrations were lower than predicted and in the northern tropical stratosphere they were slightly higher than predicted. These discrepancies may be attributable in part to inaccuracies in the predicted concentrations and in part to analytical errors in the beryllium-7 measurements, but they probably stem mainly from the mixing of air masses from different regions of the stratosphere and from the interchange of stratospheric and tropospheric air.

We have shown earlier (Figure 33) that radioactive clouds from Soviet arctic weapon tests crossed the HASP sampling corridor at 20° - 25° North latitude less than a month after their injection. With such rapid meridional transfer existing it is natural that the predicted steep meridional concentration gradients should fail to develop, and that concentrations should never reach the theoretical levels in the high latitudes. The effectiveness of the mixing process is borne out by the fluctuations in concentration from mission to mission and from month to month at individual sampling sites. The concentrations found in the tropical stratosphere probably have more nearly approximated the theoretical values because of the less effective meridional transfer in that region (clouds from weapon tests in the tropics tend to cross the sampling corridor at about the latitude of injection). Certainly air from this region does mix with that at higher latitudes, especially during the winter season, but there seems to be slightly less north-south meandering here of the air currents which comprise the zonal flow than there is in the polar regions.

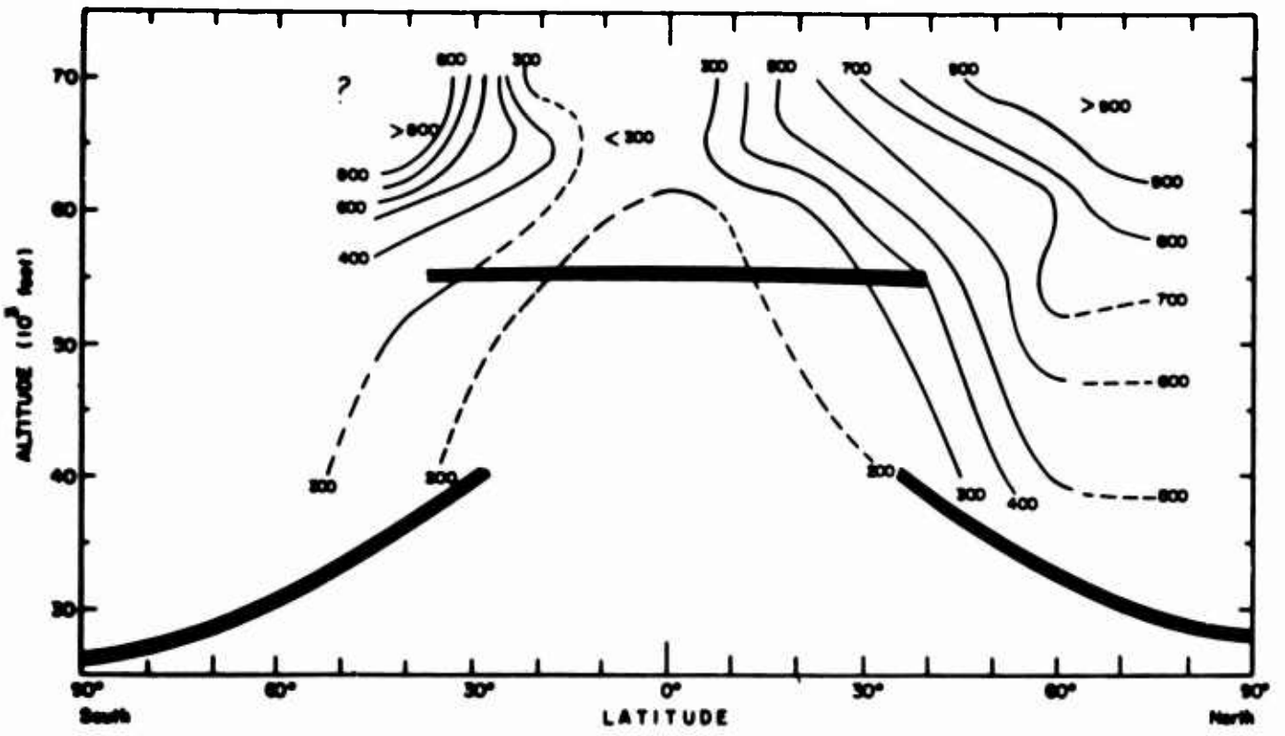


FIGURE 46 MEAN DISTRIBUTION OF BERYLLIUM-7 (dpm/1000 SCF), OCT. '59 – JUN. '60 (INCLUDING HASP AND WEATHER BUREAU DATA)

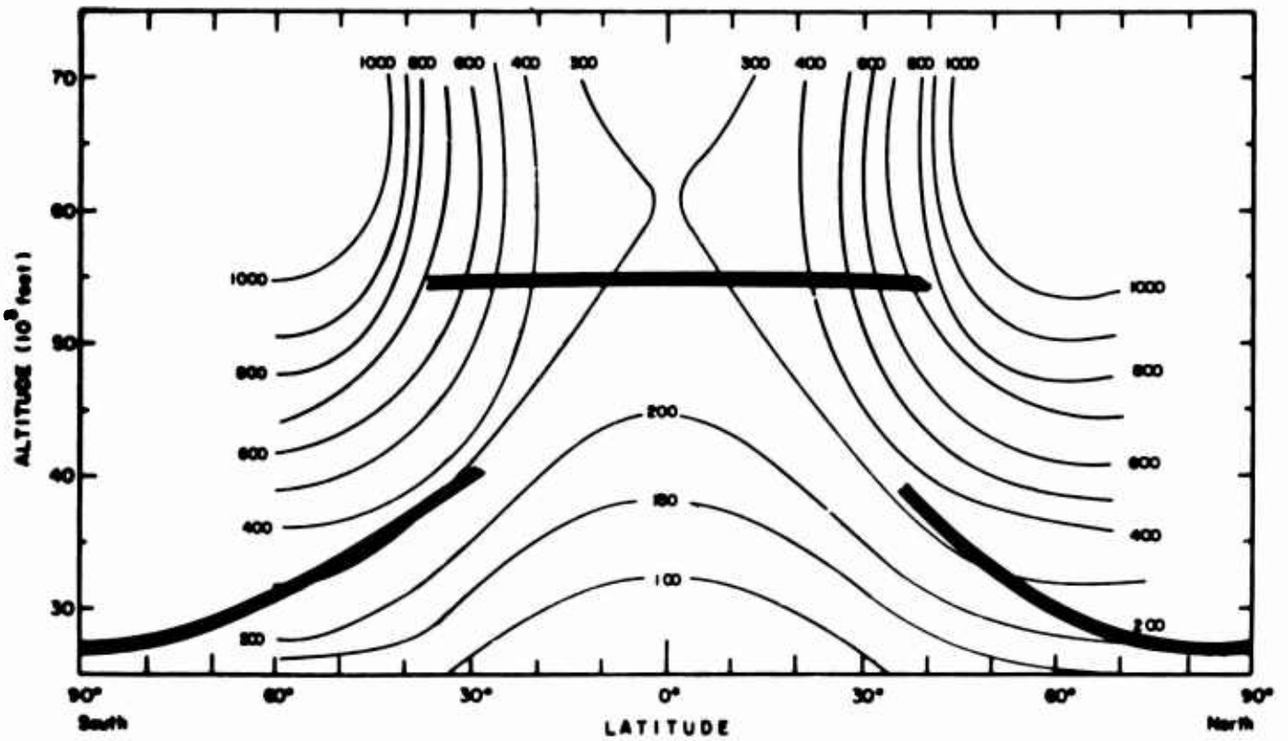


FIGURE 47 PREDICTED DISTRIBUTION OF BERYLLIUM-7 (dpm/1000 SCF) ACCORDING TO LAL

The vertical concentration gradients in the polar stratosphere are also observed to be less steep than predicted. This is doubtless the result of the relatively rapid rates of vertical mixing and of stratospheric - tropospheric exchange which exist in the polar stratosphere during the winter. It is these processes which produce the increase in rates of radioactive fallout and the increased concentrations of tropospheric ozone in the north temperate latitudes each spring. Lal[57] has suggested a method by which large scale, closed, meridional and vertical circulation patterns can be studied by the distortion of the equilibrium isopleths. The data presented here fail to reveal any flow patterns of this type. If any such exist, they must have a period which is long compared to the half life of beryllium-7. Seasonal fluctuations in mixing rates and fluctuations in cosmic ray production rates as well as other inhomogenieties may tend to blot out the departures from equilibrium. Certainly the limited data presented here cannot be expected to produce anything but a gross picture of the true average distribution. While these beryllium-7 measurements are not sufficient to permit reliable calculation of the rates of mixing and exchange, they do provide additional evidence for the importance of turbulent exchange rather than meridional circulation of the stratosphere for the transfer of stratospheric radioactivity. If the subsidence of air from high altitudes in the polar regions were an important phenomenon, as most theories of meridional circulation postulate, air with high beryllium-7 activities should have been carried down into the lower polar stratosphere during the winter of 1959-1960, and the observed concentrations there (Figure 46) should have been higher, not lower, than the predicted concentrations (Figure 47)

Phosphorous-32

Phosphorous-32 is found in the atmosphere as a result of the interaction of high energy cosmic rays on argon. Lal[57] has summarized the production rates to

be expected in the atmosphere for both beryllium-7 and phosphorous-32 (as well as other cosmic ray produced nuclides). While the absolute rate of production of these two isotopes is strongly dependent upon altitude and latitude, the ratio of the production rates expressed as Be-7/P-32 atoms per unit volume per unit time may be fairly constant in time from place to place. Lal has calculated this ratio to be about 100 atoms/atom. Since the half life of phosphorous-32 (14.3 days) is smaller than that of beryllium-7 by a factor of 3.7, then the still air, infinite bombardment concentration ratio would be 370. Lal has suggested that stratospheric air should show this latter ratio. If a parcel of this air were to be brought into the troposphere, where the absolute production rates are considerably lower, then the ratio would initially increase with a doubling life of 19 days due to the faster decay rate of phosphorous-32. A ratio as high as 1000 might be reached in 70 days or so before it gradually returned to the equilibrium ratio of 370 within one year due to production in the troposphere at lower absolute levels. Tropospheric air recently washed by heavy rains might be expected to have a concentration ratio near the production ratio of 100.

Phosphorous-32 and beryllium-7 have been reported in Indian rains from 1956 through 1959[52,53]. In reporting these data Lal and Rama have made the simplifying assumption that little stratospheric input of these nuclides occurs and that they are of tropospheric origin. This is based on the fact that stratospheric residence times are long compared to the half-lives of the isotopes. That this is a somewhat oversimplified picture, at least for higher latitudes, is evident in the seasonally dependent beryllium-7 data reported by Gustafson. On the other hand the calculated annual deposition using the above assumption agrees with the observed annual deposition for beryllium-7 within 50% and for phosphorous-32 within a factor of 2. Considering the limited data, its variability, and the inherent uncertainties in the

calculation, the agreement is quite good.

The average ratio of Be-7/P-32 in Indian rains has shown annual differences (Table XIV) which may be partially explained by the presence of some bomb produced phosphorous-32, especially in 1958. In addition, the production rate ratio may not have been constant from year to year although this seems unlikely. Finally, annual differences in rainfall, air mass history and other meteorological variables may have introduced the differences noted. In 1959 when essentially no detectable bomb produced phosphorous-32 could have been present, the ratios in Bombay rain ranged between 127 and 1150. These are close to the values described above respectively for recently cleaned tropospheric air and ex-stratospheric air which has been aged a few months in the troposphere. A certain amount of caution must be exercised in applying the results of sampling rainfall in one place such as Bombay to other latitudes or meteorological regimes. In addition, rainfall measurements may suffer from fractionation effects which will introduce anomalies in the data.

Table XIV

Bombay Rains (atoms/ml)

Year	Be-7	P-32	Ratio
1956	4200 ± 600	31 ± 7	135
1957	3400 ± 600	26 ± 6	131
1958	4300 ± 800	43 ± 8	100
1959	2900 ± 500	12 ± 2	242

A number of HASP samples have been subjected to analysis for phosphorous-32 as well as beryllium-7. Results of these measurements are shown in Table XV. While the amount of data available is quite limited in extent it appears that the ratio

Table XV

Stratospheric Concentrations of Beryllium-7 and Phosphorus-32

Sample	Collection Date	Approximate Latitude	Approximate Altitude (Kilofeet)	atoms P^{32} / g of air	atoms Be^{7} / atoms P^{32}
EN.3	10 May 60	70°N	64	10.9	224
EN.11	17 May 60	70°N	63	14.0	226
EN.2	10 May 60	70°N	60	9.12	242
EN.10	17 May 60	70°N	60	10.9	305
EN.1	10 May 60	70°N	50	7.06	229
A.18	20 Apr 60	60°N	65	10.9	185
A.2	5 Apr 60	60°N	60	10.9	231
A.26	27 Apr 60	60°N	60	8.19	237
A.7	7 Apr 60	60°N	45	5.60	244
A.29	3 May 60	60°N	40	6.20	202
A.13	14 Apr 60	40°N	70	9.65	227
A.22	26 Apr 60	40°N	70	9.21	237
*A.12	12 Apr 60	40°N	55	3.78	134
A.21	21 Apr 60	40°N	55	4.07	256
A.30	3 May 60	40°N	40	5.33	252
A.14	14 Apr 60	20°N	70	4.37	319
**A.23	26 Apr 60	20°N	70	12.4	161
3964	10 June 60	20°N	65	6.78	256
*3962	10 June 60	20°N	60	6.16	125
A.6	7 Apr 60	5°N	70	2.64	333
A.31	5 May 60	5°N	70	2.69	290
***3911	23 May 60	40°S	66	4.90	546
3930	31 May 60	40°S	65	9.73	232
3913	23 May 60	40°S	60	8.66	194
3934	31 May 60	40°S	60	4.97	290

* Abnormally low Be-7
** Abnormally high P-32
*** Abnormally low P-32

in the high equatorial region reaches 80% of the equilibrium value predicted by Lal.* At 60° N and 70°N there is a slight suggestion of a negative gradient which would be expected if there had been a downward movement at these latitudes a few months before. It is seen that the average stratospheric ratio of 252 ± 25 compares well with the average of 242 measured in Bombay rain in 1959.

Lead-210

Lead-210 (Radium-D) is a daughter product of radon-222, a noble gas formed by the decay of radium-226. Since radon-222 has a half life of only 3.82 days, little escapes from the oceans; consequently it is exhaled into the atmosphere mainly from exposed land masses. The lead-210 which is formed in the atmosphere decays according to the following scheme:

$$\underset{\text{(Ra D)}}{\text{Pb-210}} \xrightarrow{22y} \underset{\text{(Ra E)}}{\text{Bi-210}} \xrightarrow{5d} \underset{\text{(Ra F)}}{\text{Po-210}} \xrightarrow{138d} \underset{\text{(Ra G)}}{\text{Pb-206}} \quad \text{(Stable)} \qquad (14)$$

A number of investigators have reported concentrations of radon daughter products in the atmosphere[37,58-60] and biosphere [61-63]. The results of these and other investigations show that lead-210 and its daughter products form a substantial fraction of the alpha activity in plants and land animals. The source appears to be "fallout" of this particulate matter scavenged from the air by rainfall rather than by plant uptake from the soil[62,63]. Another possible source of lead-210 is neutron bombardment of the heavy elements bismuth-209 and lead-208 with the following reactions:

$$\text{Bi-209 (2n,p) Pb-210} \qquad (15)$$

$$\text{Pb-208 (2n,}\gamma\text{) Pb-210} \qquad (16)$$

*A low value of 85 for the Be-7/P-32 ratio is found in one of the Kodaikanal rains of 1959, so it may be that the production ratio of 100 predicted by Lal is 15% or so too high.

Measurements of lead-210 in air filters collected at altitudes up to 48,000 feet and in rainfall collected at the surface have been made in the U. K. In reporting these results Burton[59] has shown that between 1954 and 1956 there was a positive gradient in the troposphere (7×10^{-3} dpm/Kg of air at sea level with an increase of $\sim 10^{-3}$ dpm/Kg/1000 ft) which increased in the stratosphere ($\sim 4 \times 10^{-3}$ dpm/Kg/1000 ft). His conclusion was that lead-210 in the lower troposphere was continually being washed out in rainfall but that in the upper troposphere it had a greater residence time and could consequently reach a higher equilibrium concentration. The gradient in the stratosphere was found to be comparable to that of fission products but the explanation of higher stratospheric concentrations based on "Brewer-Dobson" circulation has certain difficulties. Primarily this would require the tropospheric concentration levels just below the tropical tropopause to be higher than those just below the polar tropopause. To produce these concentrations would require either a proportionately larger effective land mass in the equatorial regions or proportionately less atmospheric scavenging in equatorial rains when compared to higher latitude regions. Neither situation seems likely.

Telegadas[37] has reported concentrations of lead-210 in the springtime troposphere of 1960 over North America from 30° N to 56° N. In these data there was a very slight tendency towards higher concentrations at 40° N in the troposphere than at 30° N or 45° N. There was a more noticeable vertical gradient, however. Table XVI shows these average concentrations in dpm/1000 scf and dpm/Kg at various altitudes. The figures in parentheses are the number of samples used in each average value. The concentrations for 1954-1956 in the U. K. as reported by Burton are shown for comparison.

Table XVI

Lead-210 in the Atmosphere

Altitude	1960 U.S. dpm/1000 scf*		1960 U.S. dpm/Kgx10^{-3}	1954-56 U.K. dpm/Kgx10^{-3}
50	0.29	(7)	8.5	~ 80
45	0.32	(16)	9.3	50
40	0.38	(50)	11.0	38
35	0.43	(32)	12.7	34
30	0.51	(16)	14.8	30
25	0.43	(3)	12.5	26
20	0.56	(2)	16.2	21
15	0.29	(1)	8.5	18
10	--		--	14
5	--		--	10
0	--		--	7

(*For Pb-210 in air 1 dpm/1000 scf = 28.8x10^{-3}dpm/Kg = 35.4x10^{-3}dpm/m^3)

It is evident from the table that there has been a marked change especially in stratospheric concentrations in the four years that have elapsed. Concentrations have been reduced tenfold in the stratosphere and about twofold in the troposphere. As a result, the gradient in the stratosphere has become negative and the maximum concentration appears to be at about 30,000 feet. It is possible that some of the difference noted in the troposphere is due to the fact that the U. S. measurements were made during the spring while the U. K. measurements were made predominately in the summer. The increased scavenging from spring rains may have reduced the air concentrations. In addition, the U. S. measurements were "downwind" of the Pacific Ocean which could have reduced these values more than the Atlantic Ocean reduces the U. K. values. Consequently the differences in the tropospheric values may not reflect a substantial change due to the passage of time. Burton has calculated from rain water concentrations that the lead-210 fallout rate in middle latitudes is approximately 10 dpm/m^2/day. This compares favorably with the production rate of 13.6 dpm/m^2/day (a mean radon exhalation rate of 2.8×10^1 dpm/m^2/day assuming 40% of the Northern Hemisphere is land). It is noticed that the activity ratio in surface air and rain water is about 9×10^{-3} dpm/m^3 of air per 5 dpm/liter of water or equivalent activity in 1 liter of water and 550 m^3 of air. Similar equivalent activity ratios have been noted in surface air and rainfall concentrations of fission products in Norway[36] during 1959-60 where the ratio varied from about 500 m^3/liter in the summer to about 1200 m^3/liter in the winter. The possibility does exist that the differences between the U.S. and U. K. data shown in Table XVI are more apparent than real since no accurate intercalibration of the two sampling systems has been undertaken.

A number of lead-210 measurements were made in filter papers collected in the stratosphere during May 1960 (Table VI). Figure 18 shows the distribution of lead-210 in the stratosphere from these data. Tungsten-181 data collected at the same time are also shown for comparison. In both the lead-210 and tungsten-181 distributions there is a suggestion of a positive gradient above the equatorial tropopause and a negative gradient above the Northern tropopause. The distributions of the two isotopes in the Southern stratosphere are also similar. Since the tungsten distribution is due to stratospheric injections from the HARDTACK weapons test series, similarities noted here suggest that at least some of the lead-210 was also injected into the tropical stratosphere by nuclear weapons testing (but not necessarily during HARDTACK). The tenfold drop in stratospheric concentration in 1½ years noted above implies a mean residence time of about 2 years assuming all the lead-210 is from weapons testing prior to 1955. If half of the stratospheric concentration in 1960 is due to natural lead-210, then a mean residence time of 1.5 years is implied. The average stratospheric concentration of lead-210 shown in Figure 18 is 0.3 dpm/1000 scf or a total stratospheric burden of 3.6 kilocurie (0.22 moles or 16 grams). It would have been difficult to detect artificially produced lead-210 in surface measurements since it would be masked by the fallout of natural lead-210. This latter amounts to about 10 kilograms per annum worldwide while bomb produced lead-210 fallout probably could not have exceeded a few percent of that.

It should be pointed out that the data discussed above are rather limited and, as a consequence, the conclusions to be drawn from them are somewhat conjectural. Undoubtedly a considerable bit of the uncertainty as to the sources of the stratospheric lead-210 can be eliminated by continued sampling and by measuring the lead-210 content of some old HASP samples. Both of these steps are being taken. Preliminary results of measurements of lead-210 in older HASP samples (mid-1958)

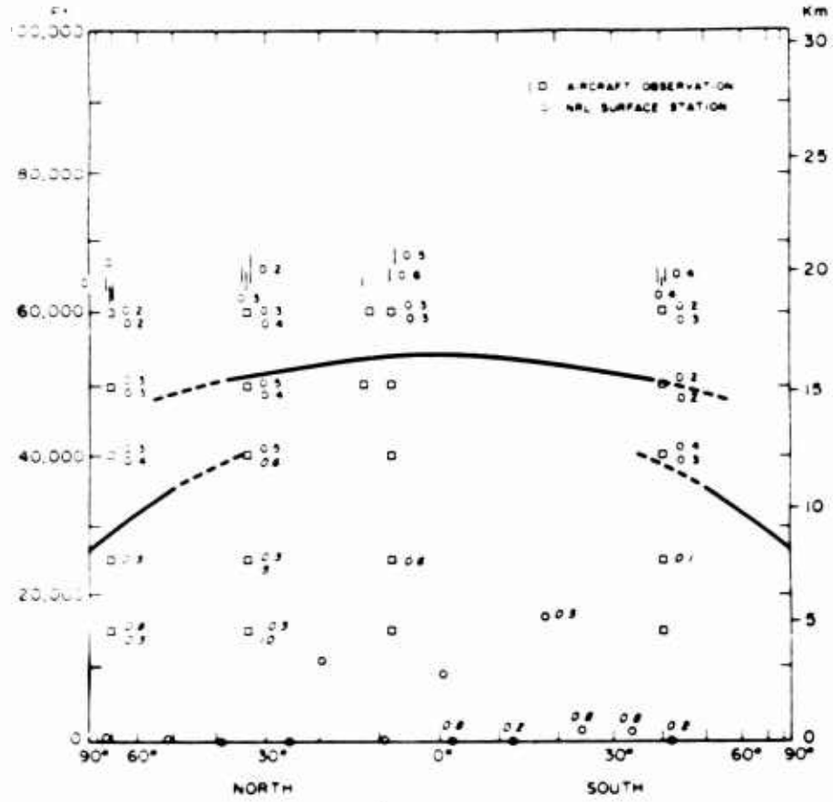


FIGURE 48a Pb^{210} d/m/1000 scf

MAY 1960

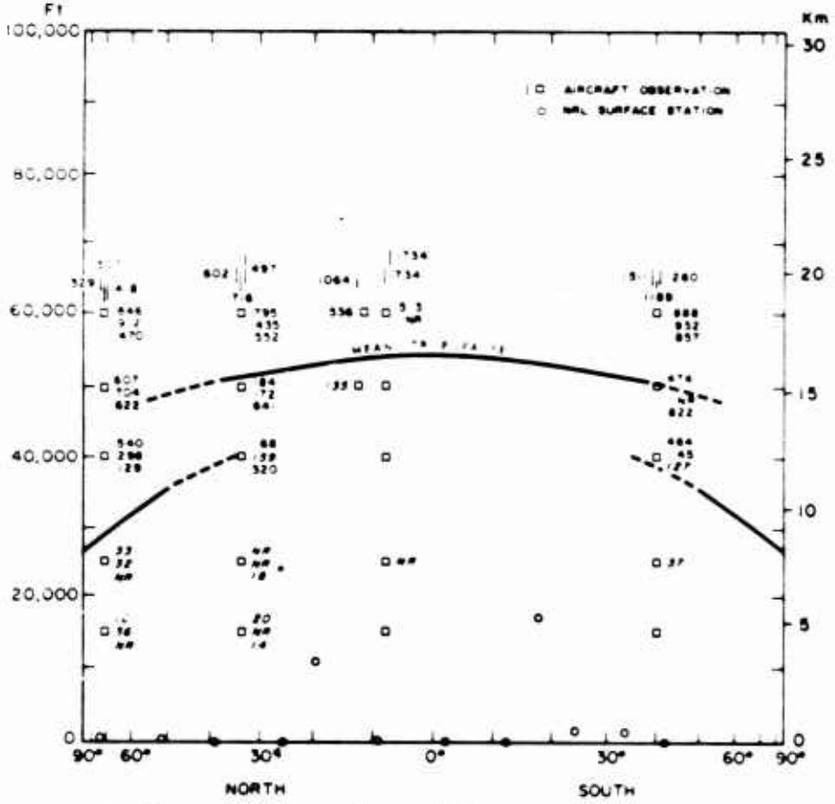


FIGURE 48b W^{185} d/m/1000 scf (corrected for decay to Aug 15, 1958)

MAY 1960

show a uniform concentration of about 0.3 dpm/1000 scf (not unlike the mid-1960 values) with no tendency to increase with increases of strontium-90. If any of this lead-210 has originated in nuclear weapons tests, it obviously predates the HARDTACK series. Until further information can be gained as to the source of the lead-210 observed in the stratosphere, caution should be exercised in interpreting radon daughter product measurements as applied to atmospheric movements.

CHAPTER VIII

FALLOUT FROM TEAK AND ORANGE

Introduction

One of the objectives of the HASP program has been the determination of the fate of fission products placed in the highest regions of the atmosphere. During the HARDTACK series of nuclear tests two weapons of megaton proportions were launched by rockets at Johnston Island in the Pacific and were detonated at high altitude. The debris from these shots stabilized in the highest levels of the atmosphere and have provided a unique opportunity to study the mixing mechanisms at these elevations. While no direct measurements of the residual concentrations from these bursts have been reported from altitudes above 100,000 feet, a number of measurements below this altitude have provided some direct and indirect evidence of the fate of this debris. Some of this evidence has been discussed before (Chapters IV-VII). An attempt will be made below to review this evidence and provide some conclusions that can be drawn therefrom.

Initial Distribution

Teak was fired on 1 August 1958 at an elevation of approximately 250,000 feet. Photographs made at Hawaii[61] showed that the airglow cloud rose with an initial velocity of approximately 1 km/sec to an altitude of around 300 km (1,000,000 feet). Orange was fired on 12 August 1958 at an altitude about half that of Teak. The debris from Orange did not rise as high as that of Teak nor were its electromagnetic or auroral effects as pronounced. In any event it is probable that most of the debris

from both shots initially rose above the mesophere into the ionosphere (See Fig. 49).

After stabilization of the cloud, the predominant forces acting upon the individual particles comprising the debris would be those of gravity and quasi-viscous retardation by the atmosphere. A number of calculations have been made of the fall rates of small particles in the upper atmosphere.[65-67] Using the terminal velocities calculated by Small[67] for unit density particles of varying diameter s d assuming no vertical motion of the air, a rough calculation of the time of fall from 300 kilometers has been made. The results are shown for submicron particles in Figure 50. It can be seen that within one week all but the smallest particles will have fallen into the mesophere. (Self coagulation processes during the first few days may result in substantial amounts of debris residing on particles in the 0.001 micron diameter range). It can also be seen that, except for the larger particles (0.1 micron to 1.0 micron), once a particle reaches the mesosphere it remains there for periods extending up to years. In other words in a "quiet" atmosphere the particles fall quickly into the mesosphere and are then virtually stopped there. Essentially the same result would occur even if the particles fell from a lesser or greater initial height (100 km or 1000 km for example).

In actuality the mesosphere is far from "quiet". Vertical mixing is probably promoted by the temperature lapse rate as is the case in the troposphere. Sodium vapor trails released in the mesosphere and lower ionosphere indicate the presence of considerable shear with vertical and horizontal as well as small scale eddy turbulence[68]. In mid-latitudes a strong zonal monsoon wind occurs in the mesosphere[69-71]. These seasonal reversals along with the vertical and latitudinal shearing action should promote rapid mixing of the debris from both Teak and Orange throughout the mesosphere. From a consideration of energy propagation within the

STRUCTURE OF ATMOSPHERE IN JULY

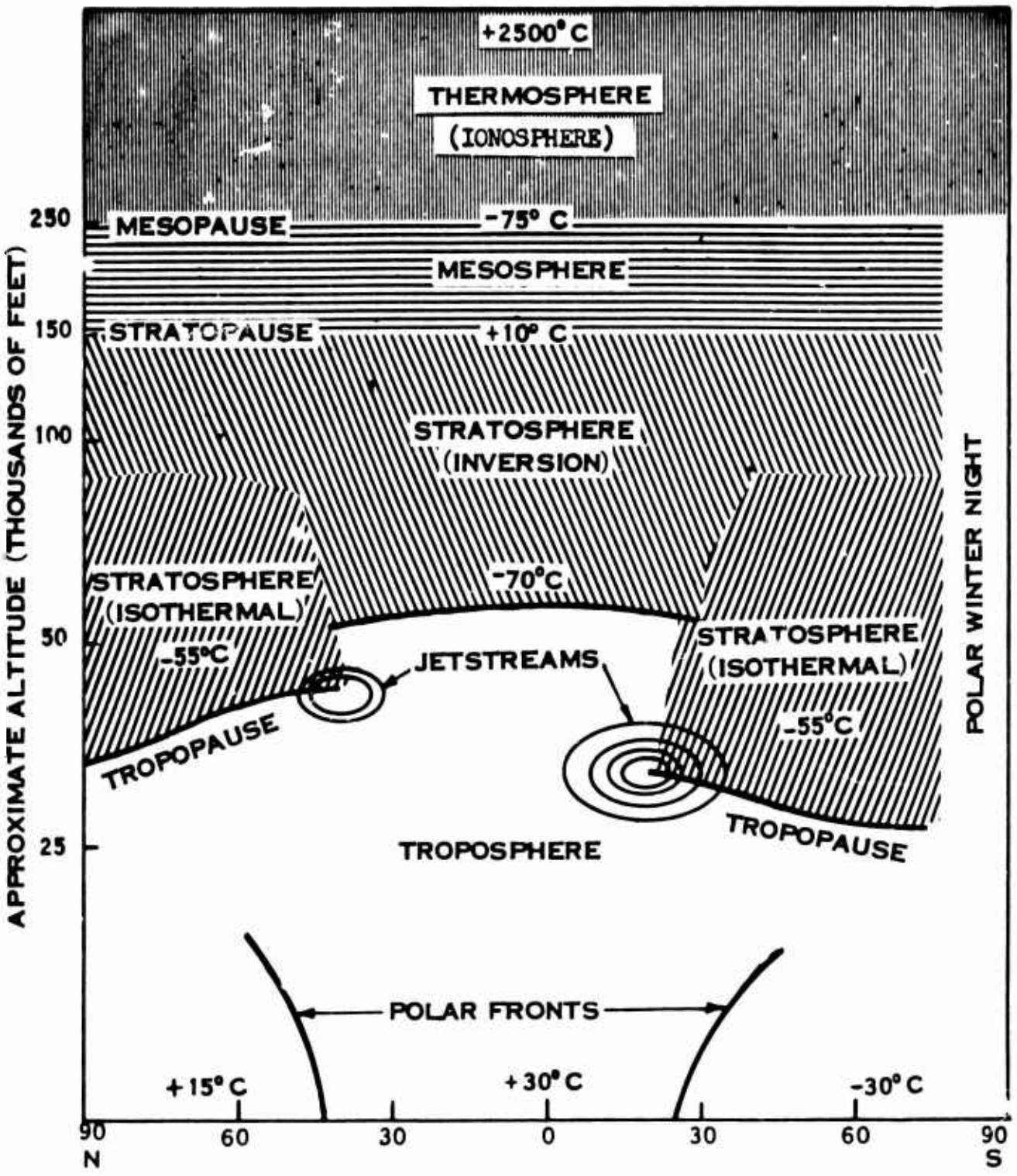


Figure 49

Altitude (thousands of feet)

stratosphere, Charney[70] concludes that the circulation in the upper stratosphere and mesosphere is, to a large extent, mechanically independent of the motion in the lower atmosphere. This conclusion would indicate, then, that the debris reaching the mesosphere would probably not mix downward to any great extent and would remain trapped in the mesosphere.

According to Glasstone[28] approximately 0.4 megacuries of strontium-90 have been injected into the mesosphere during the course of weapons testing up to 1959. If it is assumed that the major fraction of this debris came from Teak and Orange and if it is further assumed that the debris became uniformly distributed within the mesosphere, then concentrations on the order of 7000 dpm/1000 scf of strontium-90 would be expected to be found within the mesosphere. Rhodium-102 concentrations from the 3 megacurie production in Orange would be about 55,000 dpm/1000 scf corrected to shot time. This would mean that the Rh-102/Sr-90 activity ratio of mixed Teak and Orange debris corrected to Orange shot time would be about 8 to 1.

Downward Mixing

One particularly interesting feature of the polar stratosphere is that during the fall, when insolation is diminishing, a pronounced cyclonic circumpolar vortex or jet is established.[16,70-72] By early winter, the jet, which has cooled by radiation, becomes unstable and undergoes a series of "breakdowns".[71,72] These are accompanied by explosive warmings which suggest adiabatic warming of subsiding air probably due to the tilting of the isentropic surfaces with respect to the axis of rotation.[72] In the Northern hemisphere the disintegration of the vortex is usually complete a month or two before the vernal equinox while that in the Southern hemisphere occurs after the equinox. In any event it is evident that

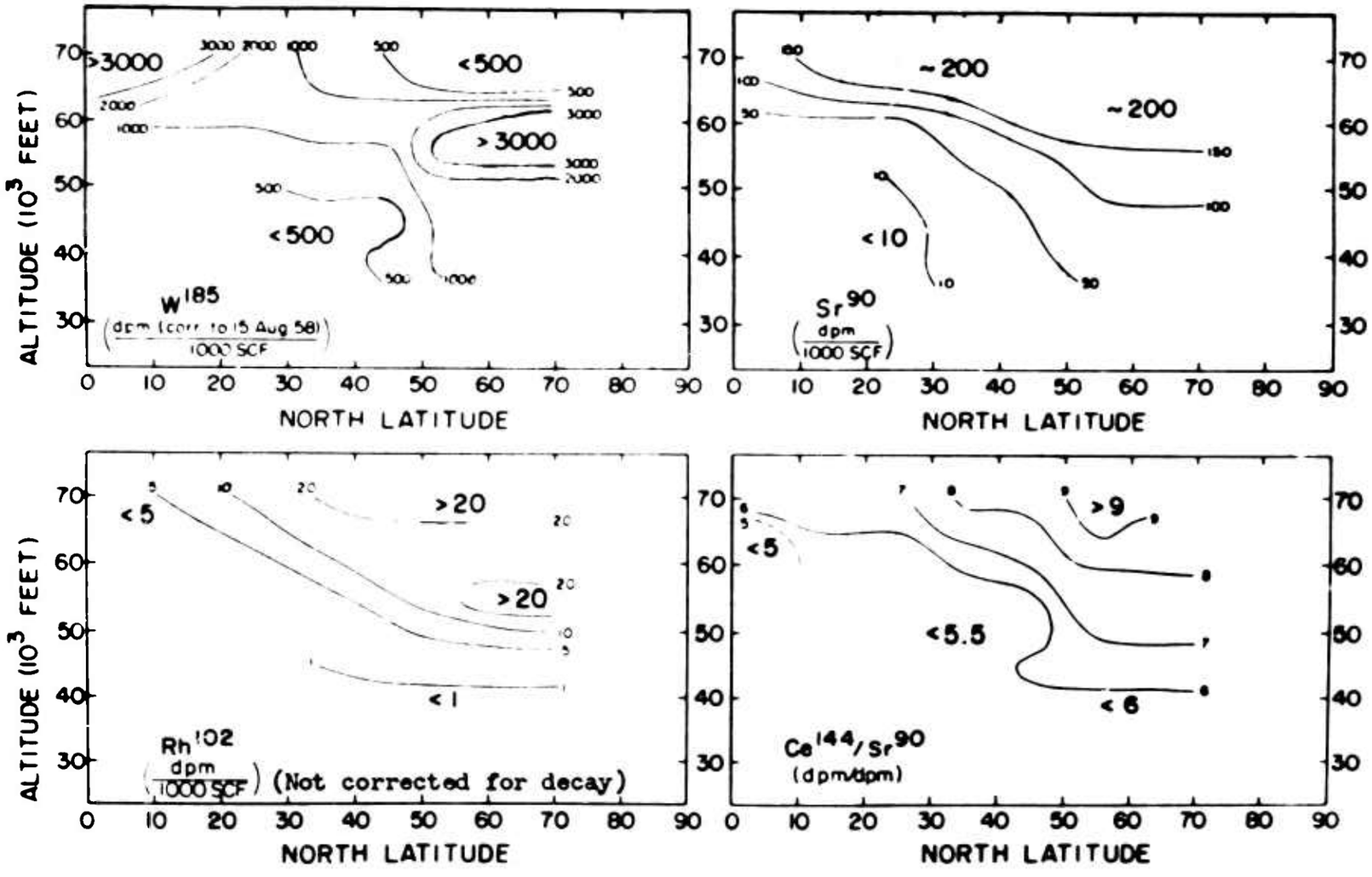
considerable interchange of polar air between the mesophere and the lower stratosphere (end even the upper troposphere) can occur during these breakdowns and may also occur when the vortex is forming in the late fall.

A crude calculation of the stratospheric concentration of strontium-90 and rhodium-102 from Teak and Orange can be made if it is assumed first that debris in the mesosphere becomes uniformly mixed and next that a localized fraction of the mixture is quickly mixed downward into the stratosphere in a vertical column centered on the pole. Since the mass of the stratosphere is about 200 times that of the mesosphere, the radioactivity concentrations described above would be diluted by that factor and one might expect to find stratospheric concentrations of about 35 dpm/1000 scf of strontium-90 and about 275 corrected dpm/1000 scf of rhodium-102. It is unlikely that the mixing in the column would be uniform down to the tropopause, consequently, higher values than those mentioned above might be found at higher altitudes. The total amount of debris removed from the mesosphere would depend upon how much of the mesosphere is involved in the polar vortex motion. As a first approximation one can say that the motion probably does not extend more than 30^{o} from the pole. Since only 13% of the mesosphere lies within 30^{o} of the pole in each hemisphere this may be a reasonable fraction to expect to be removed per annum. This would amount to a half residence time of about 5 years. Rapid mixing equatorward and downward from the polar regions with the concomitant influx of "clean" air would complete the annual cycle and allow an equivalent removal from the mesosphere during the next season.

Radiochemical Data

A detailed discussion of the radiochemical measurements of debris from Teak and Orange has been presented before (Chapters V-VI). A short review of this

discussion is appropriate here. Figure 51 shows the Northern stratospheric concentrations of four nuclides during the early spring of 1960. It can be seen that the strontium-90 distribution differs markedly from that of tungsten-185. The strontium-90 has a positive gradient at all altitudes and latitudes while that of tungsten-185 is negative in the higher latitudes and altitudes. This suggests a recent high polar source for a substantial fraction of the strontium-90. The rhodium-102 (corrected only to sampling time in this figure) has its highest concentrations at the highest latitudes and altitudes. The cerium-144 data shows that the debris is youngest where the rhodium concentrations are highest. Figure 21a showed that the strontium-90 concentrations in the Northern polar region increased from about 145 dpm/1000 scf to about 220 dpm/1000 scf during October 1959 at 70,000 feet. If half of this increase were from Teak and Orange it would be consistent with the 35 dpm/1000 scf calculated above. In addition, the apparent decrease in age of the debris shown by the cerium data (Figure 35) is consistent with an influx of strontium-90 from Teak and Orange in an amount equal to 20% to 40% of the total present. Rhodium 102 concentrations shown in Figure 38 indicate stratospheric concentrations in the 275 dpm/1000 scf range as calculated above and, in addition, delineate the stratospheric distribution of debris from Teak and Orange. By May 1960 about 0.5 megacuries of rhodium-102 had descended below 70,000 feet which again corresponds to a half residence time of about 5 years. The distribution was symmetrical about the equator and the fraction of the strontium-90 from Teak and Orange at various latitudes was respectively 25%, 10%, and 2% for the latitudes 70^{O} N, 40^{O} N and 0^{O}. More recent balloon measurements made by Ashenfelter[73] at AFCRL indicate concentrations at 85,000 feet on 27 October 1960 at Minneapolis to be about 300 corrected dpm/1000 scf of rhodium-102 and 130 dpm/1000



NUCLEAR DEBRIS IN THE STRATOSPHERE OF THE NORTHERN HEMISPHERE, MARCH – APRIL 1960

Figure 51

scf of strontium-90. This amounts to 30% of the strontium-90 being from Teak and Orange. An earlier balloon flight on 22 June 1960 at the same place showed 18% of the strontium-90 to be from Teak and Orange. Finally a small portion of the springtime increase of tritium in rainfall noted in 1960 may have come from Teak and Orange.

Conclusion

The results of the measurements described above have indirectly shown that rapid mixing probably occurs within the mesosphere. Within 6 months or so substantial amounts of debris from Teak and Orange were available in both polar regions to be mixed downward by their winter vortices. Equatorward and additional downward mixing in the stratosphere further distributed this debris where it became subject to the normal stratospheric mixing processes. The total atmospheric half residence time of debris injected into the mesosphere is probably no more than 10 years. (Mesosphere 5 $\pm$ 3 years, stratosphere 2 $\pm$ 1 year). It may be that debris stabilized just below the stratopause in the equatorial regions has an effective residence time somewhat longer than debris stabilized in the mesosphere due to slower latitudinal and vertical mixing rates, however, no evidence is available to indicate that this is so. Consequently, 10 years represents a probable upper bound on the effective total atmospheric half residence time of debris injected anywhere in the atmosphere.

One of the important implications of the above conclusion is that any debris that is released in the upper atmosphere or that strikes the top of the atmosphere from above will be subjected to this hold-up time. While this may be long enough for most of the fission products to decay almost entirely such is not the case for long lived nuclides like strontium-90 and cesium-137. Even if the half-residence time is as long as 10 years, the activity from these two nuclides will be reduced

only 20% by the time they reach the ground. Since radiation from these two nuclides accounts for about three quarters of the world-wide thirty year dose from lower atmospheric releases of fission products (Chapter XI), it appears that releases of these materials at extremely high altitudes will reduce the eventual population dose by no more than a factor of two when compared to low atmospheric releases of the same magnitude. This applies equally to any fission products which may be released outside the atmosphere and which eventually strike the top of the atmosphere.

Finally it is noted that the relative fraction of debris from Teak and Orange in the stratosphere is increasing (due to the faster removal of other debris from the lower stratosphere). It is anticipated that by 1962 over one-half of the atmospheric burden of strontium-90 will be from this source if no further testing occurs.

Chapter IX

STRATOSPHERIC MIXING PROCESSES

Introduction

In this short chapter an attempt will be made to describe the mixing and transfer processes which occur in the stratosphere as suggested by the HASP data. In addition some speculations will be offered on the location and rate of departure of nuclear debris from the stratosphere and the various mechanisms which may be involved in this transfer process. A number of features of surface fallout can be directly tied to conditions in the stratosphere and to stratosphere-troposphere interchange mechanisms. This includes the unusual latitude distribution of surface fallout and the seasonal variation in surface fallout rates and composition. Since an encyclopaedic compilation of these data and a thorough discussion of the conclusions drawn by the many investigators involved in these studies cannot be made here, only a sampling of the more pertinent studies will be presented. Additiona information on surface fallout will also be presented in Chapter X.

Vertical and Meridional Transport

Several features of the stratosphere are reflected in the HASP data. These include vertical, meridional, and seasonal movement of debris. Perhaps the most pronounced feature which relates to vertical movement is the persistance of the tungsten-185 maximum located near 70,000 feet in the tropical stratosphere (Figs. 22-31). It is evident that no steady vertical mass movement is occurring at these latitudes. During the first months after debris is stabilized in the lower equatorial stratosphere the lowest portion (55,000 - 60,000 feet) is "washed out" (probably by local fluctuations of the tropopause) and the resultant vertical gradient above the tropopause

becomes quite steep. Thereafter little of the debris appears to move across the tropical tropopause into the tropical troposphere. There appears to be some tendency of the tungsten-185 maximum to shift back and forth across the equator a few degrees seasonally, but this motion is not pronounced. Both meridional and vertical removal from the equatorial maximum can be approximated by assuming an anisotropic eddy diffusion process which will cause a net flux down the concentration gradient. It is quite apparent the meridional transport away from the equatorial regions proceeds at a rate much greater than upward movement within the equatorial regions. Using a quasi-Gaussian model of turbulent diffision (or Austauch) away from a central concentration of debris, Spar[2] has calculated both the vertical and meridional diffusion rates. In this model the half-width of the cloud grows in time according to the expression:

$$h = 1.67\sqrt{KT} \qquad (17)$$

where h is the half-width in centimeters, T is the time in seconds since injection, and K is the diffusion coefficient. The strontium-90 and tungsten-185 data suggest a value of K_y, the meridional diffusion coefficient, on the order of 10^9 cm^2/sec. The tungsten data suggests a value of K_z, the vertical diffusion coefficient, in the tropics on the order of 10^3 cm^2/sec.

The tungsten-185 data also show that meridional mixing from the tropics appears to proceed on surfaces that slope downward. The elevation of the zone of maximum tungsten-185 has lowered about 15,000 feet by the time it reaches 70^o N. No fully satisfactory explanation of this phenomenon has been offered but it may be a combination of gravitational settling especially after the debris becomes incorporated into the stratospheric sulfate layer (See Chapter III), occasional subsidence in the extratropical regions, or backflow equatorward along the downward sloping isentropic surfaces after some type of diabatic poleward movement.

Another feature of the tungsten-185 data is the saddle point in the 30° N to 50° N region (Fig. 32). This is apparently caused by poleward mixing which alternates between flow into the polar stratosphere and flow into the troposphere. This type of action could be provided either by a meandering wavelike flow through the tropopause gap or by vertical motions fore and aft of high tropospheric troughs which are associated with extra-tropical disturbances. This latter type of motion has been described by Staley[74] (see below). The action of the vertical motions Staley describes may extend some distance into the stratosphere and could also contribute to the lowering of the tungsten-185 maximum as it moves towards the poles. The apparent vertical diffusion coefficient, K_z, measured in the polar stratosphere is on the order of 10^4 cm^2/sec. In other words, vertical mixing in the polar stratosphere is more rapid than in the tropical stratosphere. It is possible that this relatively rapid vertical mixing is more pronounced in the lower polar stratosphere than in the higher polar stratosphere (say above 65,000 feet). If this were the case, then the tungsten-185 maximum could be depressed by faster downward mixing than upward mixing as the debris moves poleward.

Seasonal Trends

Several pronounced seasonal features can be noted in the HASP data. First the debris seems to be removed from the tropics into the winter hemisphere faster than it is into the summer hemisphere. This may be due to an increased meridional diffusion coefficient in the winter hemisphere compared to the summer hemisphere. Another explanation is that the volume occupied by the winter stratosphere is greater than that of the summer stratosphere and there is more "uncontaminated" air into which the debris can move. This latter situation is due to the lowering of the tropopause and equatorward movement of the tropopause gap in the winter hemisphere. In addition,

the mid-latitude jet stream moves equatorward and intensifies; which action may also contribute to increased mixing into the winter hemisphere. Surprisingly, however, the onset of the more rapid departure from the tropics seems to being at higher altitudes and then to progress downward (See Fig. 21a).

The second seasonal trend which is evident is the influx into the lower polar stratosphere of high altitude debris brought down by the polar cylonic vortex which develops during the winter. Since this action has been discussed in the previous chapter, which described the fate of debris from Teak and Orange, no further reference to it will be made here. Suffice it to say that the various seasonally dependent characteristics of the winter stratosphere are such that debris originally remote from the polar tropopause is preferentially placed nearer this tropopause in the late winter. This situation leads to the "spring rise" in fallout rate which will be described in some detail in the next chapter.

Residence Times

As the previous report (DASA 532) pointed out, the concept of a single representative stratospheric residence time is an untenable one. This concept is based on the physical notion of a steady removal from a uniformly mixed stratospheric reservoir under such conditions that the stratosphere remains uniform. As we have seen, the stratosphere has remained remarkably inhomogenious for at least two years after the last major nuclear test series (late 1958). Even in the case of debris originally stabilized in one region of the stratosphere, a single residence time probably cannot be selected to account for the departure rate for more than a few half lives and even this will show seasonal fluctuations. For instance, the tungsten-185 stabilized in the lower and middle tropical stratosphere showed a high fallout rate

initially (half residence time of about 9 months) and then a lowered fallout rate (half residence time of more than 18 months).

A number of investigators have suggested variable residence times as a function of latitude and altitude. Staley's estimates[71] have ranged from 2 months to 18 months for debris placed respectively just above the polar tropopause and in the mid-equatorial stratosphere. He makes no estimate of residence time in the high stratosphere or mesosphere. He further indicates that the residence time for some debris could be as short as a few hours if the debris were injected into the lowest stratosphere on the cold side of the jet stream associated with an upper-air frontal zone. Martell's estimates[32] have ranged from a few months for Soviet test debris to 5 to 10 years for debris at higher levels near the equator (CASTLE debris). However, both Machta[75] and Kuroda[76] have objected to Martell's treatment indicating that the spread of a factor of 60 between soviet tests and the CASTLE test is too great. On the other hand the single-valued half residence time of 0.5 $\pm$ 0.1 years which Kuroda reports based on strontium-90 concentrations in rainfall at Fayettville, Arkansas seems merely fortuitous. Libby[16] suggests a half residence time which ranges from 6 months for Soviet injections to 3½ years for high equatorial (CASTLE) injections. We have seen before (Chapter VIII) that debris injected into the mesosphere (Teak and Orange) appears to have a half residence time on the order of 5 years and that 10 years probably represents an upper limit on the half residence time of debris injected anywhere in the atmosphere (including debris which falls on the top of the atmosphere).

Even though the assignment of varying half residence times to various locations in the stratosphere may not be proper in the strictest sense, it is tempting to do so because the calculation of the resulting stratospheric and surface inventories becomes quite simple. Figure 52 shows the results of a calculation of the quarterly stratospheric inventories below 150,000 feet and the surface inventories of strontium-90

originally in the stratosphere based on half residence times of 5 months for the lower polar stratosphere, 10 months for the tropical stratosphere up to 70,000 feet, 20 months between 70,000 and 100,000 feet in the tropics, 30 months between 100,000 and 150,000 feet in the tropics, and 60 months above 150,000 feet. The strontium-90 was decayed at 2½% per annum. The vertical distribution of debris within the cloud and the amount injected into the stratosphere was calculated according to the method described in the previous report with the additional assumption that 50% of the strontium-90 was scavenged into local fallout from land surface bursts and 30% from water surface bursts. It can be seen that the soil and rainfall inventories at the surface agree reasonably well with the calculation, but that the calculated stratospheric inventory drops off after 1959 at too rapid a rate. This latter discrepancy is enhanced by downward mixing into the HASP and Ashcan sampling region of debris from Teak and Orange (which were accounted for only in the surface inventory).

Meridional Circulation Models

Besides the Feely-Spar[8] model of meridional transport of debris through turbulent diffusion processes which has been described above, two other schemes involving organized circulation patterns to account for meridional movement within the stratosphere have been suggested. The first of these is the Brewer-Dobson model.[11,15] This model suggests that tropospheric air moves upward across the tropical tropopause and then proceeds northward and southward deep within the stratosphere escending in the polar or temperate regions and thence back into the troposphere. This model was originally evoked to account for the dryness of the polar stratosphere and the efflux of ozone from the stratosphere into the higher latitude troposphere. A number of investigators[11, 15, 19, 59, 77-79, 81] have found

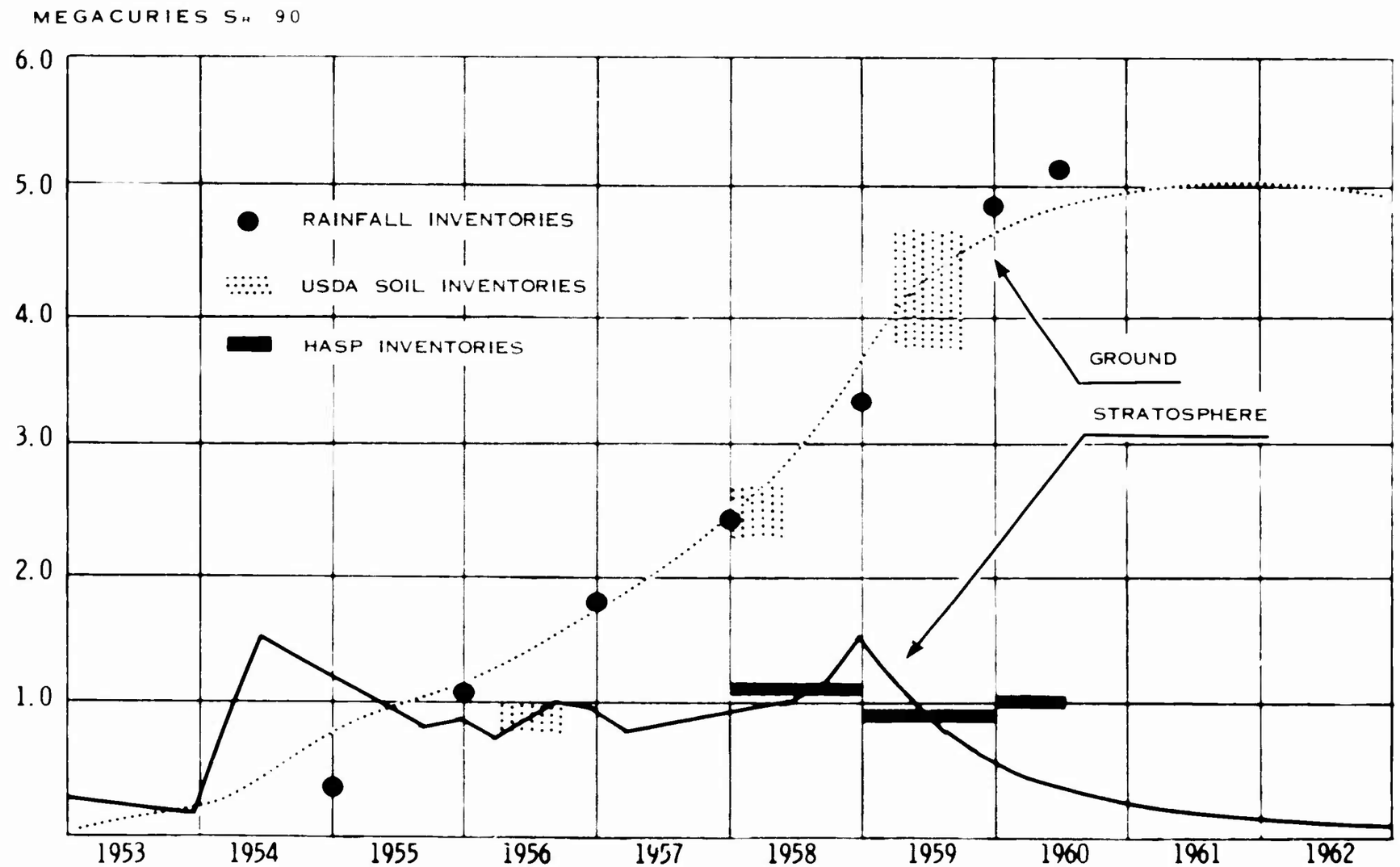


FIGURE 52 STRONTIUM-90 INVENTORY

this circulation pattern to be compatible with surface measurements of fallout material as well as stratospheric measurements of various radioactive materials. In almost every case, the Freely-Spar model can be used equally well to account for the distributions and concentrations observed. On the other hand the tungsten-185 data obtained by HASP seems to be entirely incompatible with the Brewer-Dobson model since the equatorial maximum has shown no tendency to rise. Furthermore the steep gradient noted in the polar regions for all isotopes militates against a persistent subsidence of air in this region as required by the Brewer-Dobson circulation. Except for the "cold trap" mechanism to explain the apparent low water vapor concentrations in the polar stratosphere there seems to be little evidence left to commend the Brewer-Dobson circulation pattern. Recent measurements of relatively high water vapor content at 90,000 feet[19]* suggest that the low concentrations in the lower polar stratosphere may be accounted for by eddy diffusion out of the tropical tropopause region into the polar stratosphere and the possible interference with frost point measurements by the hygroscopic sulfate layer present in the lower stratosphere.**

More recently Libby and Palmer[16] have suggested a meridional circulation cell which lies completely within the stratosphere. This pattern provides for slow poleward movement over a large region of the higher stratosphere and more rapid equatorward movement in a smaller region of the lower stratosphere with upward movement at the equator and downward movement at the poles. Since the pattern described lies almost entirely within the HASP sampling region, this type of circulation almost certainly should have been observed in the HASP data if it in fact exists. Such has not been the case. Libby[80] suggested that the tungsten-185 injected into the lower tropical stratosphere was prevented from entering this circulation pattern by being trapped in the Berson westerly winds which lay just above the tropical tropopause.

*See also Mastenbrook, H. J. and J. E. Dinger Journ Geophys Res 66 1137 (1961)

**See also Goldsmith, P. and F. Brown Nature 191, 1033 (1961)

However these winds had disappeared from the tropical stratosphere by the late summer of 1958[81,82]* and were replaced by the more general Krakatoa easterly wind regime which prevailed thereafter. This fact seems to vitiate Libby's argument somewhat. In any event, neither the strontium-90 distributions nor the tungsten-185 distributions observed by HASP tend to support the Libby-Palmer circulation model. Finally, independent support of the Feely-Spar model is provided by Newell[83] who suggests that the surface measurements of tungsten-185 originally placed in the stratosphere indicate that this material was carried poleward and downward by eddy processes rather than those of so-called mean motions.

Stratospheric-Tropospheric Interchange

A number of investigators have suggested possible modes of removal of radioactive debris from the stratosphere. Spar[2] has shown that a pure eddy diffusion across the tropopause may occur but that it is too slow to account for the short residence times observed. He has further suggested that turbulence in the tropopause gap region might produce higher horizontal diffusion coefficients and could promote turbulent transfer through the gap. Large scale horizontal meandering stream flow through the gap was also suggested. Without suggesting how the debris crossed the tropopause, Machta[85] has calculated that the net zonal removal of strontium-90 from the stratosphere during the spring of 1959 was proportional to the area of the zone i.e. the net flow per unit area north of 30° N was uniform.

Perhaps the most evident feature of the surface fallout is that it descends at a maximum rate during the spring and has a peak in the 30° - 60° latitude band. Among others, Gustafson[40] has suggested that the spring peak can be explained by the increasing volume occupied by the spring-time troposphere as the tropopause rises and as the gap region moves poleward. This action would encompass

* See also Reed, R. J., A. J. Campbell, L. A. Rasmussen, and D. G. Rogers Journ of Geophys Res 66, 813 (1961)

stratospheric air which has had sufficient time to have its radioactive content enhanced by the seasonal stratospheric phenomena described above. In addition, Gustafson attributes considerable influx to a disruption of the polar tropopause in middle latitudes during the spring months. He further points out that little stratospheric removal occurs in the late summer and fall while the polar tropopause is lowering and the gap is moving toward the equator. This is evidenced by the fact that surface air concentrations decrease rapidly during this time (apparent residence time of less than 50 days). Walton[86-88] has discussed the effect of subsidence of stratospheric air into the troposphere in the wake of cyclonic disturbances. These disturbances are prevalent in the mid-latitude region and produce heavier rainfall during the spring months. Walton's studies have shown that the tropopause can be lowered by these disturbances and it is possible that as it is reëstablished at a higher altitude some of the stratospheric debris will be incorporated into the troposphere.

In a classic study of atmospheric phenomena in the vicinity of the polar-equatorial tropopause discontinuity (tropopause "gap"), Staley[71] has reviewed some of the above notions and points out the difficulties involved in considering the tropopause to be some kind of barrier to airflow with the gap region providing a hole in this barrier. In fact his study shows that the usual definition of the tropopause becomes inappropriate in the gap region when associated with an extra-tropical disturbance. Here the discontinuity in potential vorticity appears to be the significant physical quantity. This construct is the same thing as the tropopause away from the gap region and in the gap region defines what has been described as a "folded" tropopause or tropopause "funnel" which may extend down to 800 mb (5000 feet). Considerable vertical motion of the air in the vicinity of this "funnel" has

been noted. In the lower stratosphere subsidence occurs over areas as large as $2x10^{4}$ Km^2 with velocities as great at 10 cm/sec in the frontal zone usually found on the southwest side of the trough aloft. Within 24 hours some stratospheric air can be brought down along isentropic surfaces (adiabatically) to within 5000 feet of the ground. It also appears that diabatic incorporation of stratospheric air into the troposphere can take place. Rising air is found on the northeast side of the trough aloft. Trajectories show that most of the rising air is not air that had just subsided and vice-versa. In other words, the circulation is not closed in the immediate vicinity of the disturbance and there is a net interchange of air. Since the tropospheric residence time of fallout debris is short, it is to be expected that the radioactivity will be removed from the air which descended from the stratosphere prior to its return to the stratosphere from the troposphere. This would result in a net outflow of debris from the stratosphere. Assuming that there are 5 such storms operating in each hemisphere at all times, Staley has shown that a net quantity of air equal to half the mass of the stratosphere is exchanged in about 18 months. His conclusion concerning the mass flow out of the stratosphere associated with extratropical disturbances of moderate intensity in middle and high latitudes as being quantitatively important for the residence time of debris injected into the stratosphere seems entirely justified if the stratospheric behavior described above is taken into account. Further it provides considerable insight into the mechanism of interchange in the "gap" region. It is not to be concluded, however, that Staley's mechanism is the only one which produces egress of radioactive debris from the stratosphere, since the other mechanisms described above doubtless are real. The relative importance of these mechanisms cannot be fully evaluated

at the present time; moreover, their effectiveness probably would be strongly dependent upon the precise conditions and location of injection of a particular cloud of debris into the stratosphere.

Chapter X

SURFACE FALLOUT

Introduction

While the HASP program has contributed little direct data on surface fallout distributions, since its major aim was to delineate the role played by the stratosphere in the world-wide distribution of fallout, it was hoped tnat this program would contribute to an understanding of the surface distributions observed. While it will be impossible here to produce an encyclopaedic survey of the many fallout measurement programs which are presently being conducted and which have been conducted in the past, it will be instructive to note some of the features which have been observed in a few of the more extensive collection networks.

Surface measurements generally fall into three categories, namely, measurements of activity in surface air, surface rain, and soil (and sea water). Surface air measurements generally reflect the amount of material available for removal by rainfall. A single station may be representative of a relatively large area and will reflect to some extent the seasonal influxes from the stratosphere tempered by the removal of debris by recent rains. Surface rainfall measurements generally reflect the rate of fallout at the collection station and may not be as representative of as large an area as are the surface air measurements due to the localized nature of rainfall. Total inventories can be evaluated by integrating the rainfall collections taking into account the variations from place to place of specific isotopic concentrations and rainfall amounts. Except in areas which experience little rainfall, dry fallout is a relatively unimportant factor since rainfall is believed to be the primary scavenging agent for fallout debris over most of the globe.

Soil measurements provide a direct method of determining the total surface burden of radioactive fallout. However, this technique has a number of difficulties in practice which include: problems in radiochemical extraction, problems in accounting for rainfall variations from place to place, and problems in accounting for radioactive decay of the various nuclides.

In the sections which follow, several topics will be discussed including; the latitudinal and seasonal variation in fallout rates from various sources, total inventories both surface and atmospheric, fallout from the French tests, fallout from the Nevada Test Site, and certain characteristics of concentrations in rain water and the soil.

Distribution of Surface Fallout

One feature of surface fallout which has been almost universally observed[31,33,36,40,76,79,84,87,89-96] is the fact that the rate of fallout in the spring is greater than in the fall by a large factor. One of the most striking displays of this seasonal variation is provided by measurements of cesium-137 in surface air collected at the Argonne National Laboratory near Chicago. Figure 53, provided by Gustafson,[40]* shows that there has been an increase in the concentration each spring since the first thermonuclear detonation. A number of explanations of this phenomena have been described in previous chapters and it seems that it is caused primarily by seasonal changes of the meteorological conditions in the upper atmosphere. It has also been suggested that "seasonal" variations in nuclear testing cycles have produced the periodicity noted. Certainly these testing cycles affect the spring fallout rate but it seems reasonable to believe that they relate more to the quantity

* See Gustafson, P. F., S. S. Brar, and M. A. Kerrigan Science 133, 460 (1961)

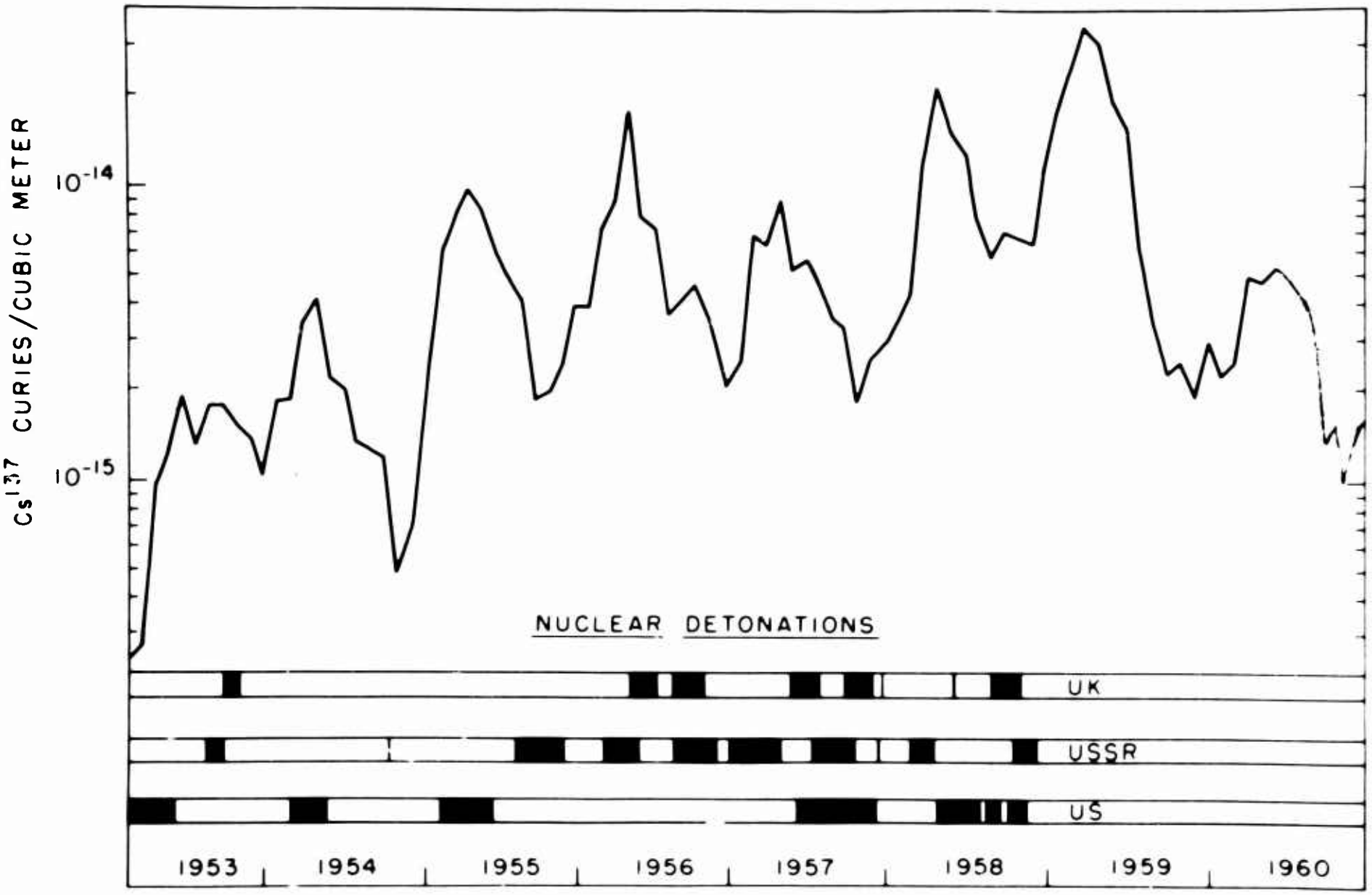


FIGURE 53 SEASONAL VARIATIONS OF CESIUM-137 CONCENTRATIONS IN CHICAGO AIR.

of fallout produced each year than to the yearly frequency observed. Hvinden[93,95] has suggested that annual variations in rainfall may account for the seasonal variations in the concentrations noted in surface air and rain water. He has shown that the increase in rainfall during the spring and summer tends to depress the air concentrations which later build up when the rainfall diminishes. Hvinden concludes that stratospheric fallout rates may be fairly constant in time. While the seasonal factor in rainfall no doubt enhances the seasonal fallout effects, it is difficult to attribute the tenfold fluctuation in air concentrations to the twofold fluctuation in mean rainfall.

An extensive surface air sampling network has been operated along the 80th meridian by the U. S. Naval Research Laboratory since 1956.[31,33] Several of the features of this network have been described in previous chapters. The network has shown that, in general, the greatest concentrations of radioactive material occur in both mid-latitude regions. In addition it has shown that concentrations in the Southern hemisphere have been consistantly lower and the debris has been older than in the Northern hemisphere. These differences have been due mainly to the high latitude Soviet tests. The 1959 spring peak in the Northern hemisphere was well documented and it was shown that the bulk of this peak came from the fall 1958 Soviet test. After May 1959, the surface air concentrations in the Northern hemisphere dropped rapidly which indicated that little debris was entering the troposphere from the stratosphere. By late 1959, most of the debris that was measured at the surface appeared to be of HARDTACK origin.

Gustafson's[40] extensive investigation of air samples collected at ANL sheds additional light on some of the surface fallout features after mid-1958. Figure 54 shows the concentrations of tungsten-181, cesium-137, and rhodium-102 measured by

gamma-ray spectroscopy in the air samples collected. The tungsten-181 is presumed to come from the HARDTACK series alone and the rhodium-102 from Orange. Using these isotopic measurements as well as others, Gustafson was able to reconstruct the fraction of the cesium-137 observed which could be attributed to various sources. A multiple isotopic analysis similar to that described in Chapter VI was employed along with a meteorological construct to eliminate the contribution of pre-HARDTACK debris. The results are shown in Figure 55. It is seen that about 80% of the 1959 spring peak came from the Soviet test of late 1958. No further debris from this source was seen after late 1959. The spring peak of 1960 was mainly from HARDTACK and included an increasing amount from Teak and Orange. The apparent stratospheric residence time has increased due to the removal of the lower altitude elements. Gustafson indicates that roughly 50% of the cesium-137 found in the surface air during the fall of 1960 was pre-HARDTACK in origin.

Similar results to those of Gustafson concerning the fallout from the Soviet test series have been reported by a number of investigators. Walton[90] concludes from tungsten-185 and strontium-89,-90 measurements in New Jersey rain that 70% to 80% of the strontium-90 in the 1959 spring peak came from the Soviet test. A true seasonal variation of the equatorial debris is also observed in the tungsten-185 collections at Westwood from the HARDTACK series. Using a method similar to that of Gustafson, Edvarson[92] concludes that only 55% of the cesium-137 in the 1959 spring peak in Sweden was attributable to the Soviet series and that the residence time of this material was about 6 months. On the basis of tungsten-185 measurements in the world-wide AEC pot (rainfall) collection program, Hardy[34] concludes that no more than 20% of the strontium-90 in the 1959 spring peak can be assigned to

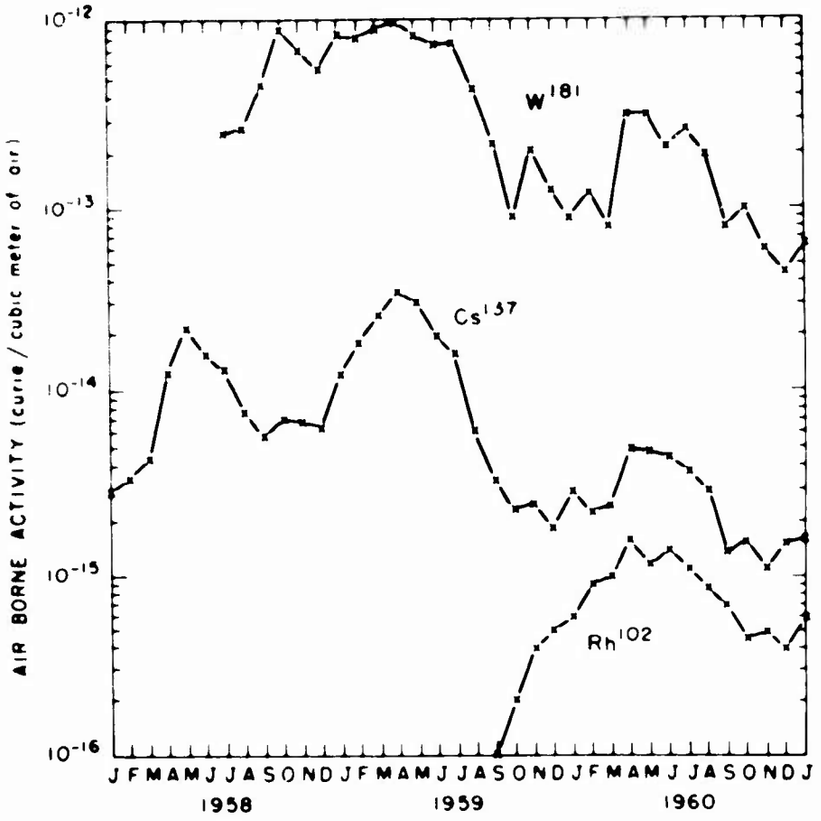


FIGURE 54 CONCENTRATION OF CS-137, W-181 AND RH-102 IN SURFACE AIR AT AN

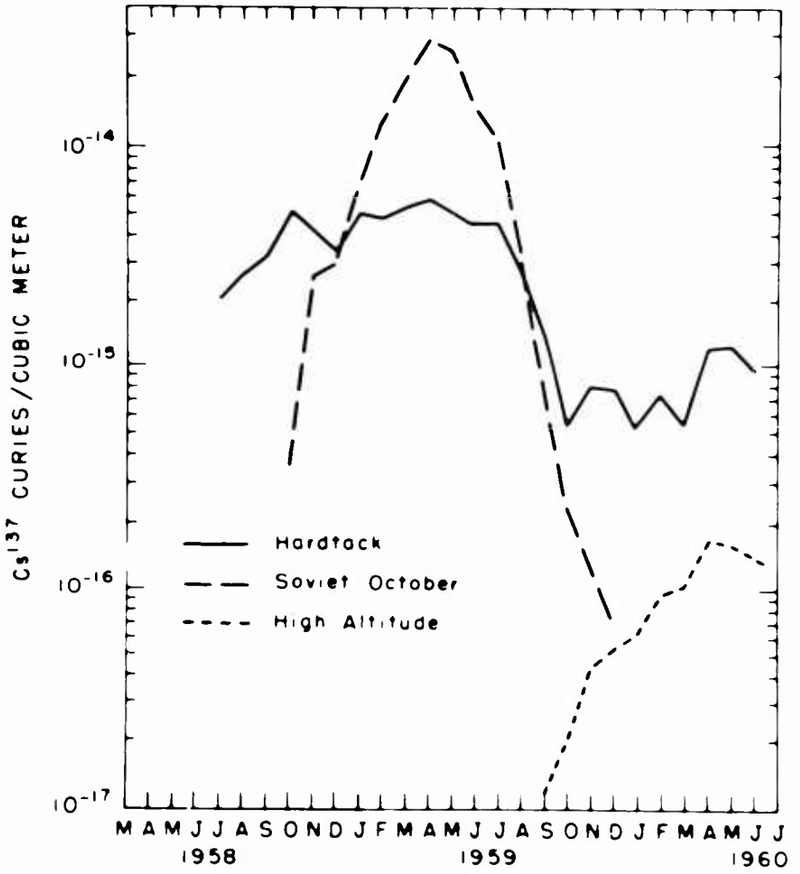


FIGURE 55 SOURCES OF CS-137 IN SURFACE AIR AT ANL

the HARDTACK series. Bleichrodt[89]* has reported the strontium-90 spring peak in 1959 in rainfall over Holland to be composed of as much as 80% to 90% Soviet debris. He also notes the rapid decrease in concentration after May 1959 (initial half-time somewhat more than one month). He attributes this to a sudden reduction in the rate of input of radioactive dust from the stratosphere. Bleichrodt also observes a peak in the spring of 1960 which he attributes to meteorological factors. Kuroda[77] has reported that 60% of the 1959 spring peak in Arkansas rain can be attributed to Soviet debris. The 1960 spring peak is also apparent in his data.[76]

A number of studies of fallout debris in rainfall in the United States have been undertaken. Some of the results obtained by Walton's[87,88] study of New Jersey rain will be mentioned here. The most evident feature that is noted in the New Jersey rain is the seasonal fluctuation in concentration. From September 1959 through July 1960 the concentration of strontium-90 could be approximated by a steady fallout of 0.08 mc/mi^2/in with a spring excess of about 0.15 mc/mi^2/in. Over 40% of the total deposit for this period came from the excess. Over the years, the rate of accumulation of strontium-90 at Westwood, N. J. and New York City calculated from rainfall measurements has paralleled the rate of departure from the stratosphere. Table XVII shows the deposit each year up to July. It can be seen that the deposit prior to 1954 was low and that between mid-1951 and mid-1958 the annual rate was fairly constant (~10 mc/mi^2/yr). From mid-1958 to mid-1959 the rate more than doubled and thereafter dropped off to a low value. The net deposit of 83.5 mc/mi^2 reduces to 78.5 mc/mi^2 on 1 July 1960 if radioactive decay is taken into account. The changes in rate noted here compare with those shown previously

* See also Bleichrodt, J. F., Joh. Blok, and E. R. van Abkoude, Journal of Geophysical Research 66, 2183 (1961)

in Figure 52.

Table XVII

Incremental and Net Deposits of

Strontium-90 in the Westwood, N. J.

Area Since 1954 (mc/mi^2)

To Mid-Year	Increment	Net (undecayed)
1954	4.2	4.2
1955	9.5	13.7
1956	10.9	24.6
1957	11.5	36.1
1958	13.1	49.2
1959	27.1	76.3
1960	7.2	83.5

Measurements of the Ce-144/Sr-90 ratio in Westwood rains can be compared to those collected elsewhere. This is done in Figure 56 for rain collected at Westwood, N. J., Pittsburgh, Pa., Houston, Texas, and Richmond, California. In addition the lower stratospheric concentrations obtained by HASP and the rainfall measurements at Rijswijk, Holland are shown for comparison. It can be seen that there is considerable difference between the stations. Part of this may be due to differences in laboratory calibrations but this should be a minor factor (and should be absent in the Westwood-HASP comparison). It is evident in the figure that the January 1960 U. S. readings are all higher than the HASP readings. This may be due to the last increments of Soviet debris being deposited while the HASP measurements in the lower stratosphere reflect the incursion of HARDTACK debris. No support for the contention that the January rainfall contains much Soviet debris can be provided by the Chicago air measurements, however (see Fig 55). After January the overall average U. S. rainfall measurements drop to about that value which would be predicted

from lower stratospheric measurements but the individual U. S. station averages show large variations. The rainfall measurements at Rijswijk[101] agree remarkably well with the HASP measurements, however.

Walton[88] has investigated the notion that the low values at Westwood are caused by resuspension of strontium-90 previously deposited. He shows convincingly that this cannot be the case. One clue as to why the various U. S. stations show so much variability from month to month and from one another is supplied by solubility studies on various nuclides in rainfall. Salter[42] has reported that the cerium-144 in one Pittsburgh rain sampled was about 1% water soluble while the strontium-90 fraction was about 50% water soluble. This disparity may well lead to fractionation between the nuclides during the course of their incorporation into rain and eventual deposition. In addition, this disparity places a premium on careful collection and analysis techniques. Salter concludes that large fluctuations in the Ce-144/Sr-90 ratio in precipitation may be expected, depending upon the history of the precipitating elements, and the relative quantity of particulate matter included. It should be noted also that, like the Ce-144/Sr-90 ratio, the Cs-137/Sr-90 ratios in rainfall and soil samples collected at a number of sites[36,101] have often differed markedly from the theoretical ratio. Measurements in stratospheric samples collected by HASP lead to the conclusion that these differences are engendered after departure from the stratosphere since the ratios in the HASP samples are close to the theoretical value.

Walton[88] has reported that dry fallout may be enhanced in cerium-144 and may show greater variability than fallout in rain water. In addition, he notes that the specific concentration of strontium-90 in the light rain following a cold frontal passage is often greater than that in the heavier rain in the frontal zone. Finally, Walton has shown rather conclusively that one cannot reconstruct soil

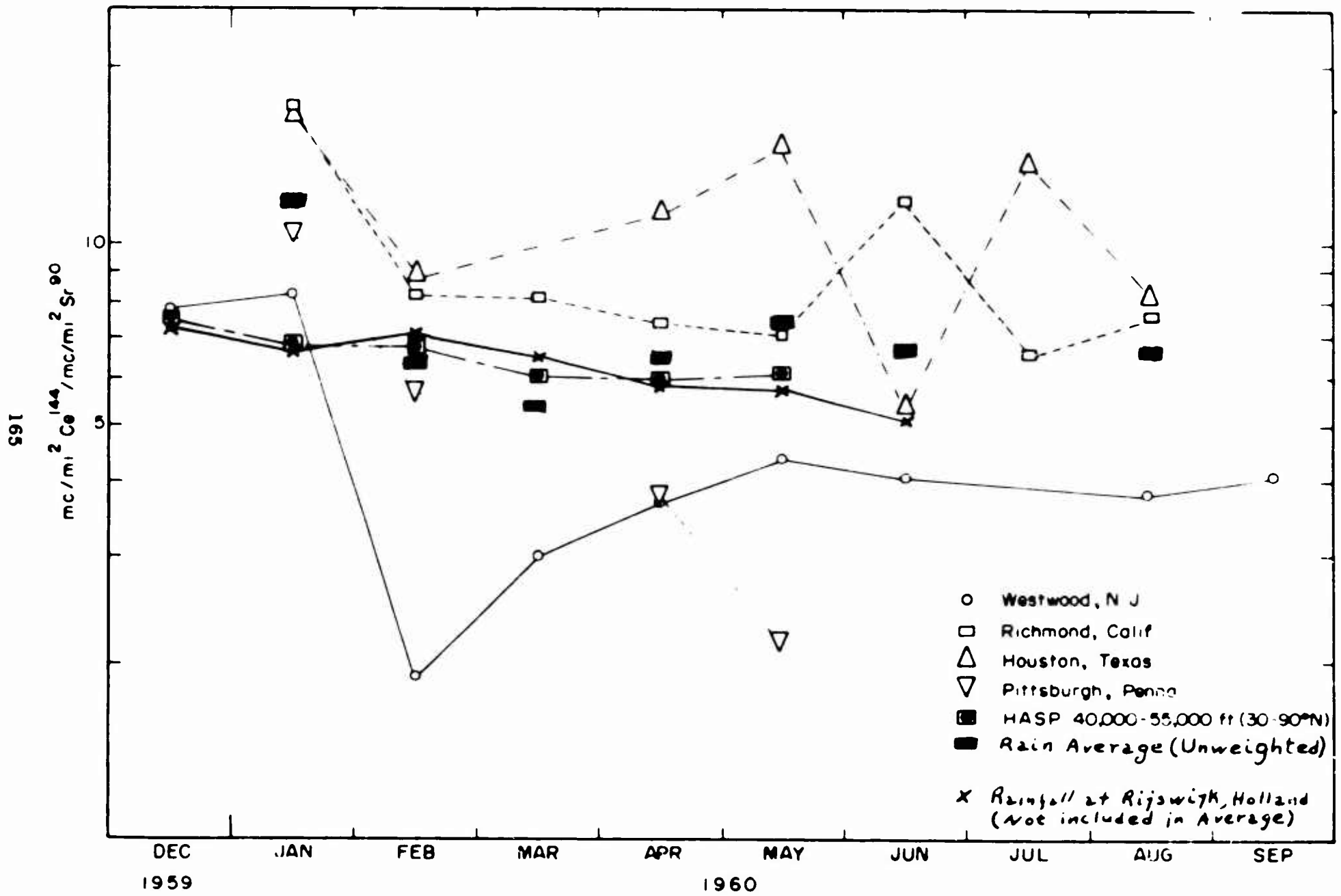


FIGURE 56 CE-144/SR-90 RATIOS IN AIR AND PRECIPITATION SAMPLES

concentrations of strontium-90 at one geographical location from rainfall concentrations at another site even when the two are at the same latitude and have the same amount of rainfall. This indicates that local meterorological conditions as well as variations in tropopause heights, elevation of clouds producing precipitation, and the history of particular air masses may affect differently the rate at which various nuclides are incorporated into rain. The fact that Rijswijk lies downwind of a large uniform region (the Atlantic Ocean) may explain why its rainfall measurements reflect so closely the lower stratospheric measurements.

The major point to be made here is that extreme caution should be used in making generalizations from rainfall collected at one geographical location.

Fallout from French Nuclear Tests

On 13 February 1960 the French detonated a device with approximately 60 to 80 kilotons yield. Since a number of months had elapsed since the last detonation, the presence of its debris in the 80° meridian surface air network was quite apparent.[97] Twenty days after detonation it had reached three-quarters of the way around the globe and the cloud had a half width at the surface of about 20° latitude. Using expression (17) above, this yields a meridional diffusion constant of 10^{10} cm^2/sec for the equatorial troposphere. The effective zonal wind (not necessarily the surface wind) would be 35 statute miles per hour. A similar passage was noted for the 27 December 1960 shot, but, even though it reached the network in one-half the time, concentrations were one-fifth to one-tenth those of the first shot.[99] A number of other investigators have reported measurements of debris from the first French detonation.[36,37,76,87,89,98,100] These all occurred in surface air and rainfall at higher latitudes than that of the explosion. Apparently the high tropospheric winds were not zonal during the course of the first circumglobal

traverse. In addition, the cloud probably was split up into a number of segments due to wind shears.

To determine what effect this detonation had on the "spring peak" of 1960, it is necessary to determine the fraction of activity (both short-lived and longer-lived) attributable to the test. A rough approximation can be made by comparing the strontium-90 produced in the French test (~6 Kilocurie) with the amount available for the spring fallout in the lower stratosphere of the Northern hemisphere. Table VII shows this to be about 330 Kilocurie. It appears then that at least 2% of the fallout could come from the French test. It is an easy matter to determine the French contribution of any isotope observed in a particular sample by measuring its strontium-89 or barium-140 content. Kuroda[76] reports that the rain sample at Fayetteville, Arkansas with the most French debris amounted to 3% of the total strontium-90. Walton[87] reports a maximum in one New Jersey rain of 6% which increased the total of that month (February 1960) by 1%. Bleichrodt[89] reports an increase in cesium-137 for the month of April to be 2.3% of the total at Rijswijk. It is seen, then, that the spring increase in fallout of longer lived isotopes is only slightly affected by the French test. On the other hand, a large fraction of the spring increase in total beta activity can be attributed to the French debris at a number of stations since there were proportionately more short lived beta emitters in the French cloud than in the older background. For instance the 1960 spring increase in cumulative fallout (mc/km^2 based on beta decay) at Kjeller, Norway reported by Hvinden[36] appears to be mainly from the French test.

Surface Inventory of Strontium-90

Two basic types of data are available for determining the total burden of strontium-90 on the surface of the earth, namely concentrations of this nuclide in

soil and in rain. Soil measurements provide the most direct measure of the total concentration. There are a number of different methods of attack which may be used to extend the sparse soil data available to represent the whole globe. Walton[102] and Alexander[103] have examined several of these methods and have detailed the strengths and weaknesses of each.* Walton's basic system is to sum over each 10^{o} latitude band the product of: the band area, A_i in mi^2; the mean deposition concentration, $\bar{f}_i$ expressed in mc/mi^2 per inch of mean annual rainfall; and the mean annual rainfall for the latitude belt, $\bar{p}_i$ in inches. The analytical expression is:

$$\text{Inventory} = \sum_{i=1}^{n} A_i . \bar{f}_i . \bar{p}_i \qquad (18)$$

One way to obtain the value of f_i for any one site is to divide the quantity of strontium-90 in a soil sample (mc/mi^2) by the amount of rainfall that fell at that site between 1953 and the time the sample was taken and then multiply by the number of years between 1953 and the time the sample was taken. Assuming that in any one latitude band the deposition concentration is independent of the mean annual rainfall (MAR) at the various sites, $\bar{f}_i$ is obtained by taking the arithmetical mean of f_i for each site. Using values of $\bar{p}_i$ determined from extensive climatological studies one can then calculate the inventory. Table XVIII shows Walton's[102] results based on the above system.

One major deficiency in the above calculation is that the assumption that the deposition concentrations, f_i, are independent of the mean annual rainfalls at various sites in any one band is probably not valid. Walton has shown that there is a negative correlation between these two variables, i.e., the soil deposition

* See also previous report (DASA 532)

TABLE XVIII

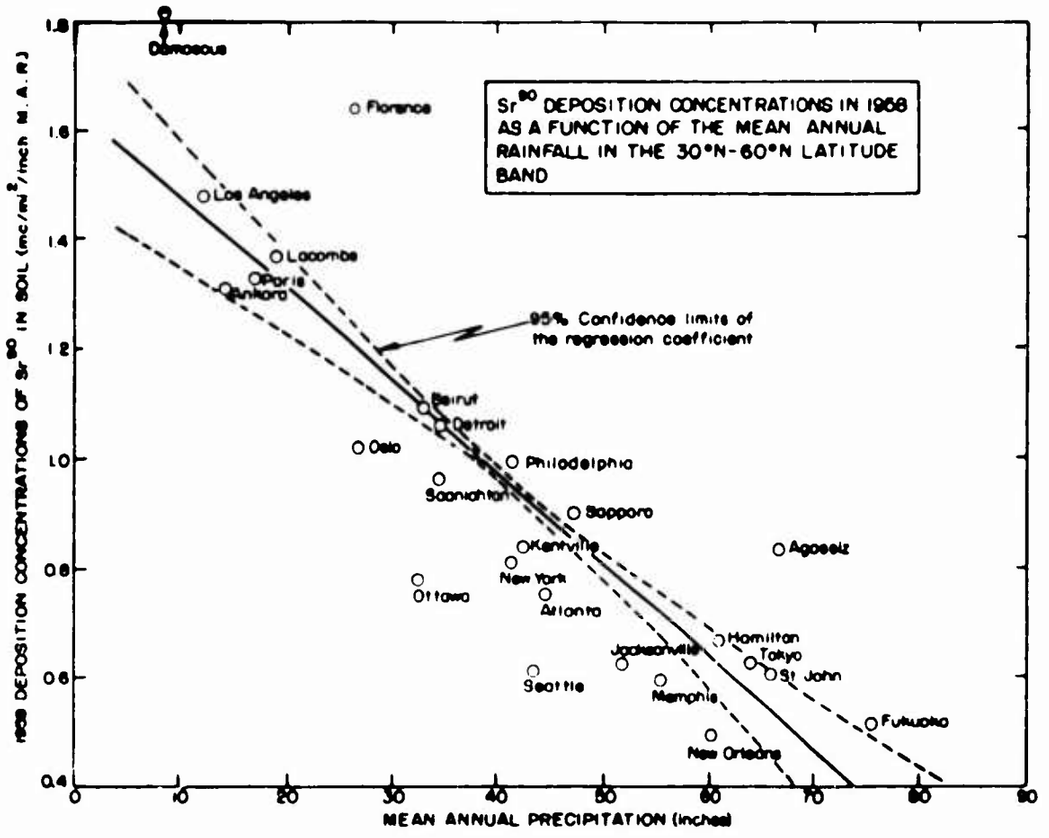
Surface Burden* of Strontium-90 on 1 July 1959

Latitude	Area ($mi^2 \times 10^{-6}$)	MAR** (inches)	Deposition Concentration (mc/mi^2/in. MAR)	Deposit (megacuries)
80° - 90°N	1.6	4	1.47	0.009
70° - 80°N	4.3	6	1.47	0.038
60° - 70°N	7.3	14	1.47	0.150
50° - 60°N	9.8	26	1.47	0.375
40° - 50°N	12.2	31	1.47	0.556
30° - 40°N	14.0	28	1.47	0.576
20° - 30°N	15.5	25	0.95	0.378
10° - 20°N	16.5	34	0.54	0.303
0° - 10°N	17.1	61	0.19	0.198
0° - 10°S	17.1	46	0.22	0.173
10° - 20°S	16.5	37	0.23	0.140
20° - 30°S	15.5	26	0.40	0.161
30° - 40°S	14.0	31	0.39	0.169
40° - 50°S	12.2	44	0.39	0.209
50° - 60°S	9.8	38	0.39	0.145
60° - 70°S	7.3	16	0.39	0.046
70° - 80°S	4.3	3	0.39	0.005
80° - 90°S	1.6	1	0.39	0.001
				3.63 MC

* Calculated from soil data.
** Mean Annual Rainfall from Möller.

concentrations in areas of low rainfall are proportionately higher than those of higher rainfall. Figure 57 shows this relation for the 30° N to 60° N latitude band for soil and rainfall up to mid-1958 and mid-1959. The deficiency described above may be overcome by using plots like those shown in Figure 57 for various latitude bands to obtain $\bar{f}_i$ directly. This is done by using the intercept on the curve corresponding to the value of $\bar{p}_i$ for that latitude band. Walton has shown that this correction will increase the soil inventory for 1 July 1959 from 3.6 megacuries to 4.3 megacuries.

Alexander[103] has used a different approach to obtain the inventory. His method is to plot the total concentration in mc/mi^2 observed in a soil sample directly upon an equal area projection of the globe which contains climatological rainfall contours. Lines of equal concentration are then drawn among the plotted points using the rainfall contours and plots such as those shown in Figure 57 as guides to interpolation. Alexander's result for mid-1959 is shown in Figure 58. The total inventory obtained by integrating the isolines with a planimeter is 4.1 megacuries. This compares well with Walton's result. One possible source of systematic error which the soil data are subject to is incomplete recovery of strontium-90 in the radiochemical processing. Alexander has shown that as much as 16% of the strontium-90 may be overlooked in the soil samples. By far, the greatest uncertainties, however, are the amount and distribution of precipitation over the globe since 1953, and the quantities of fallout in the oceans. Limited evidence[47,103] indicates that the specific activity of strontium-90 in rainfall over the oceans is greater than over the inland regions where most of the soil collections are made. In addition, dry fallout which has amounted to 15% of the total in some areas[47,104] will distort some of the analytical corrections which are made on the basis of rainfall.


Damascus
Florence
Los Angeles
Lacombe
Paris
Ankara
Beirut
Detroit
Oslo
Philadelphia
Saanichton
Sapporo
Kentville
New York
Ottawa
Atlanta
Agassiz
Hamilton
Tokyo
St John
Jacksonville
Seattle
Memphis
Fukuoka
New Orleans
95% Confidence limits of the regression coefficient
Sr90 DEPOSITION CONCENTRATIONS IN 1958 AS A FUNCTION OF THE MEAN ANNUAL RAINFALL IN THE 30°N-60°N LATITUDE BAND
1958 DEPOSITION CONCENTRATIONS OF Sr90 IN SOIL (mc/mi2/inch M.A.R.)
MEAN ANNUAL PRECIPITATION (inches)

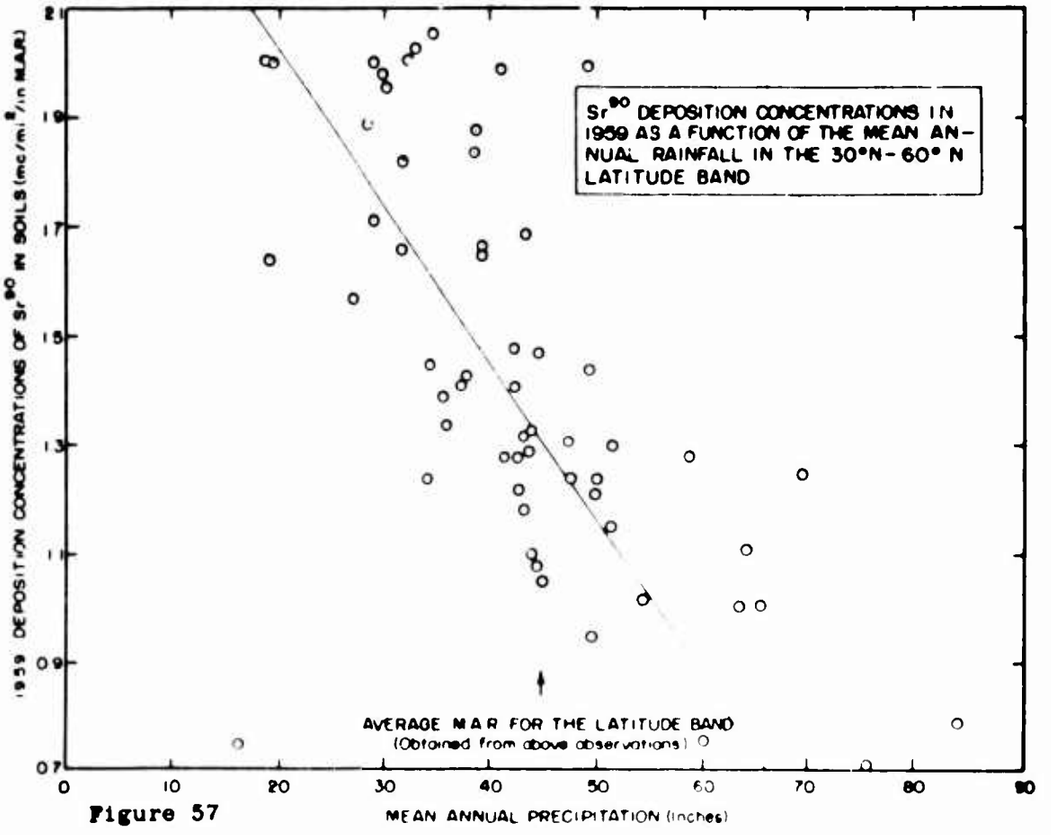

Sr90 DEPOSITION CONCENTRATIONS IN 1959 AS A FUNCTION OF THE MEAN ANNUAL RAINFALL IN THE 30°N-60° N LATITUDE BAND
AVERAGE MAR FOR THE LATITUDE BAND
(Obtained from above observations)
1959 DEPOSITION CONCENTRATIONS OF Sr90 IN SOILS (mc/mi2/in MAR)
MEAN ANNUAL PRECIPITATION (inches)

Figure 57

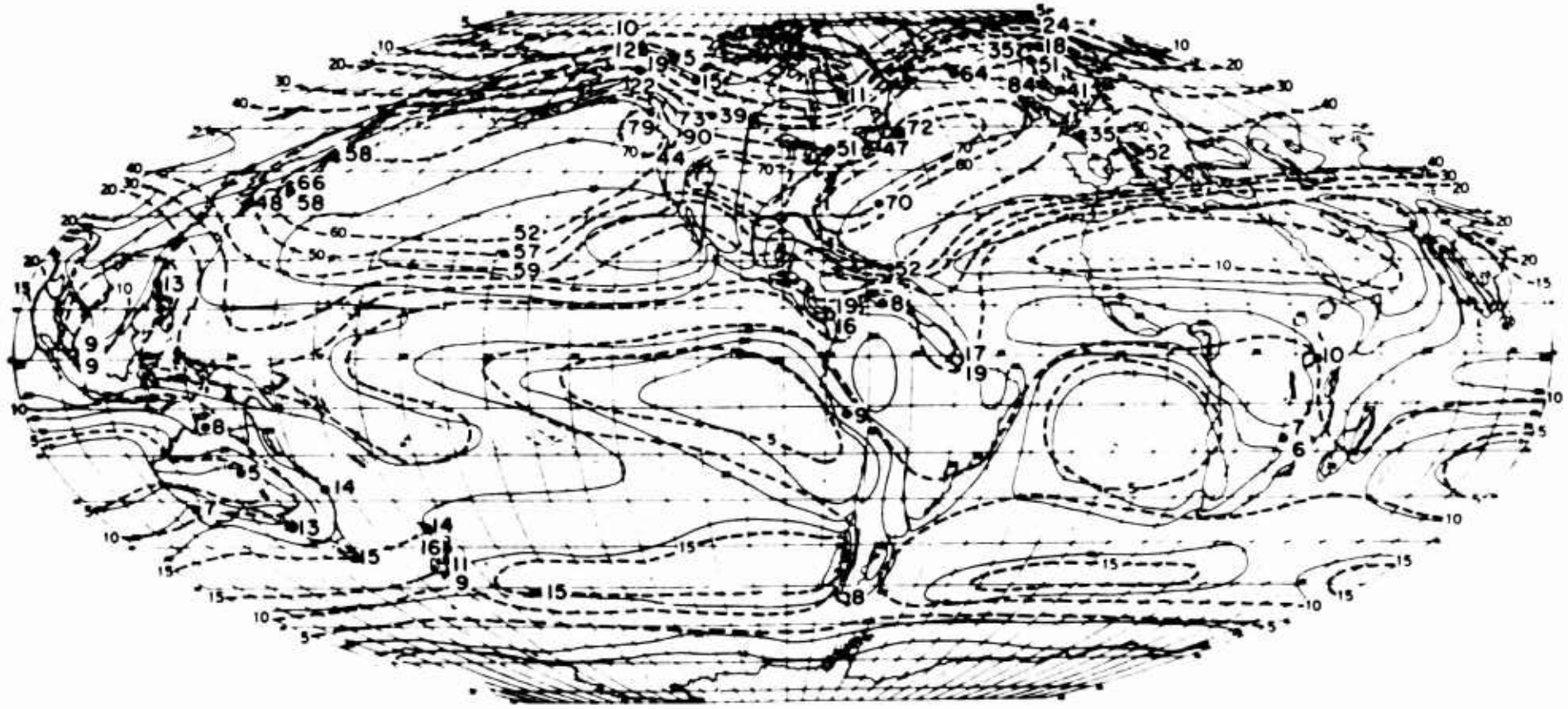


FIGURE 58 1959 SOIL STRONTIUM-90 AND MEAN ANNUAL PRECIPITATION.

The overall error in the soil inventories calculated above may be as much as 1 megacurie.

Another somewhat independent source of data for computing the surface burden of strontium-90 is that provided by measuring concentrations of this nuclide in rainfall. Telegadas[105] has calculated quarterly increments by two methods. One calculation is made by summing the product of the average deposition in mc/mi^2 at a number of stations in any one latitude band times the area of the latitude band. Another calculation is made in a similar manner except the average deposition is corrected to the amount which might be expected if each station were to experience the climatological mean rainfall. Using a similar approach, Walton[102] has calculated the values of $\bar{f}_i$ based on rainfall for each quarter since 1953. These values are shown in Table XIX. Immediately evident in this table are the differences between the Northern and Southern hemispheres and the spring (second quarter) peak in the Northern hemisphere. In the Southern hemisphere the "spring" peak appears to be more of a "late winter" (third quarter) peak. Using these figures and applying them against expression (18) above, Walton calculates the sum of the quarterly global increments to be 3.89 megacuries on 1 July 1959. If each quarter is subjected to radioactive decay ($2\frac{1}{2}\%$ per annum) then the inventory on 1 July 1959 reduces to about 3.6 megacuries which is essentially the same as the soil inventory of 3.63 megacuries calculated under the same premises. At the bottom of Table XIX is a calculation of the "rainfall" burden of strontium-90 at the end of each year from 1954 to 1959. The method of calculation used is that described above. In addition, a somewhat arbitrary increase of 15% is added to each annual total to account for the negative correlation between f_i and p_i which was noted in the inventory calculations based on soil data. It should be pointed out that the same

TABLE XIX

Idealized Concentrations (mc Sr^{90}/mi^2/inch) of Strontium-90 in Precipitation for the Period 1954 - 1959

Quarter	South Latitude				North Latitude			
	90-30	30-20	20-10	10-0	0-10	10-20	20-30	30-90
1954								
1	0.03	0.01	0.01	0.01	0.01	0.02	0.06	0.11
2	0.03	0.01	0.01	0.01	0.01	0.03	0.11	0.16
3	0.05	0.03	0.01	0.01	0.01	0.03	0.06	0.09
4	0.05	0.03	0.02	0.02	0.02	0.03	0.08	0.10
1955								
1	0.06	0.02	0.02	0.02	0.02	0.04	0.13	0.23
2	0.06	0.06	0.02	0.02	0.03	0.09	0.21	0.36
3	0.07	0.12	0.03	0.03	0.03	0.07	0.16	0.24
4	0.07	0.12	0.01	0.01	0.02	0.04	0.08	0.10
1956								
1	0.07	0.03	0.02	0.02	0.02	0.06	0.19	0.33
2	0.07	0.06	0.02	0.02	0.03	0.12	0.32	0.47
3	0.09	0.06	0.02	0.02	0.03	0.05	0.12	0.18
4	0.09	0.05	0.02	0.02	0.03	0.06	0.11	0.19
1957								
1	0.09	0.02	0.02	0.02	0.02	0.05	0.11	0.21
2	0.09	0.04	0.03	0.03	0.03	0.08	0.20	0.30
3	0.06	0.06	0.03	0.03	0.02	0.05	0.13	0.20
4	0.09	0.06	0.03	0.03	0.02	0.05	0.12	0.16
1958								
1	0.07	0.02	0.02	0.02	0.02	0.05	0.18	0.25
2	0.04	0.04	0.03	0.03	0.03	0.09	0.28	0.58
3	0.07	0.10	0.03	0.03	0.03	0.07	0.16	0.30
4	0.11	0.10	0.03	0.03	0.03	0.07	0.16	0.35
1959								
1	0.14	0.08	0.03	0.08	0.16	0.57	0.62	0.82
2	0.09	0.10	0.09	0.04	0.03	0.26	0.33	1.21
3	0.07	0.10	0.04	0.04	0.03	0.05	0.11	0.24
4	0.14	0.08	0.02	0.05	0.01	0.04	0.10	0.10

Year End Global Surface Inventory*
(Megacuries of Strontium-90)

1954	1955	1956	1957	1958	1959
0.37	1.06	1.81	2.11	3.34	1.85

*Based on summation of above data using Mollers MAR with a 15% increase due to negative correlation between f_i and p_i (from soil data)

difficulties in analytical treatment and the same sources of error that apply to "soil" inventories apply equally to "rainfall" inventories. Soil sampling has the advantage that correction for decay is unnecessary and furthermore, if weathering is negligible over a period of several years, it makes little difference when the samples are collected. Rainfall samples are not as subject to radiochemical loss as are soil samples, but they only provide increments for the periods in which continuous collections are made.

Material Balance of Strontium-90

An estimate of the material balance of strontium-90 can be made from the above data. Based on reasonable climatological and soil data, Walton's best estimate of the surface burden on 1 July 1959 is about 1.3 megacuries. When added to the atmospheric burden estimated in Chapter V of about 1.3 megacuries in mid-1959 we note that a total of about 5.6 megacuries is distributed world-wide on 1 July 1959. If this strontium-90 had not been subject to radioactive decay since its formation, it would have totaled 5.9 megacuries (5% or 2 years average decay). This compares favorably with previous estimates by Glasstone[28] and Libby[16] of the net stratospheric injection of 5.9, 6.2 and 6.5 megacuries respectively. The remainder of the 9.2 megacuries originally produced has apparently descended in local and tropospheric fallout into areas away from sampling network stations. About 0.1 megacurie descended in local and tropospheric fallout from the Nevada Test Site (See below). As a consequence soil and rainfall samples immediately downwind of Nevada were not used to determine the average latitude concentrations. It may be that some of the high specific values noted in Canada, Alaska and Northern Europe are from tropospheric fallout from the Soviet test sites and bias the surface inventory slightly if only the stratospheric component of the surface inventory is being sought.

Figure 52 in the previous chapter showed that by late 1961 the total surface inventory was expected to reach a maximum of about 5 megacuries. This situation would prevail when the rate of decay of the total strontium-90 on the ground just equalled the rate of fallout from the stratosphere. Figure 59 shows the estimated average latitudinal distribution on the surface when this occurs. Mean annual rainfall for each 10^{o} latitude band is also shown.

Tropospheric Fallout from Nevada

A substantial fraction of the fallout experienced in the Plains States in the U. S. is from tropospheric fallout from the Nevada Test Site. As a consequence one cannot use the values of Sr-90 in soil and rainfall in these areas to calculate world-wide fallout as they will tend to bias the calculation in the 30^{o} N to 50^{o} N latitude band. An estimate of the fraction of the fallout from Nevada which has fallen on the U. S. can be obtained by reversing the world-wide fallout calculation, however. Walton[87] has predicted the world-wide fallout to be expected in the U. S. by using Figure 57 and the known rainfall at various stations in the U. S. since 1953. By subtracting these values from the observed soil values an "excess" at each station can be determined. Figure 60 shows these excesses at a number of stations. Isolines have been drawn through the points down to 10 mc/mi^2. A large "hot spot" centered on Nevada is immediately evident. Integration around the contours down to the +10 mc/mi^2 isoline produces an inventory of 40.9 kilocuries or about 40% of the total strontium-90 produced from the 970 kilotons of fission products released in Nevada. Usually firing of the tests in Nevada occurred when the upper wind trajectories were to the northeast. Consequently, most of the remaining fraction of the Nevada fallout apparently has descended over Canada and the Atlantic Ocean outside of soil sampling locations, although some undoubtedly appeared in local

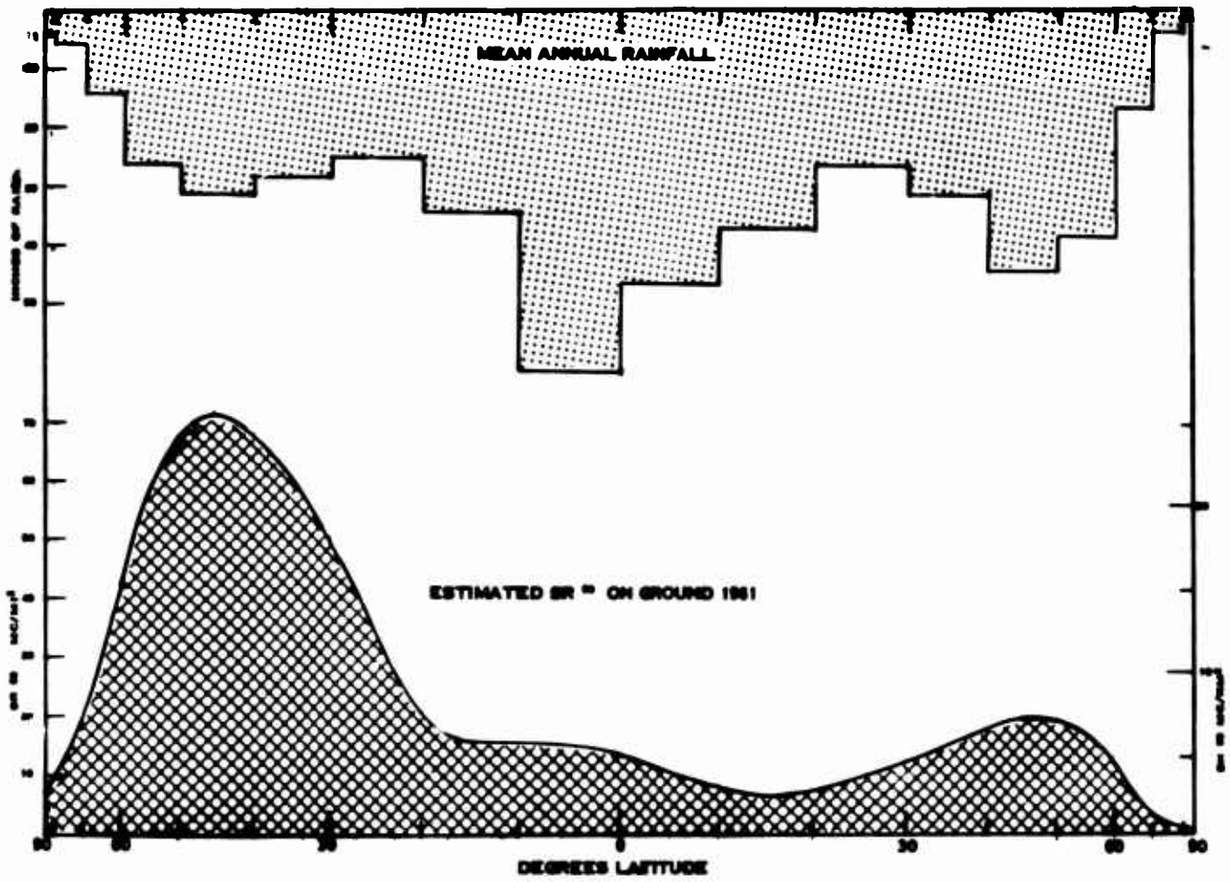


FIGURE 59 WORLDWIDE DISTRIBUTION OF STRONTIUM 90– AND RAIN

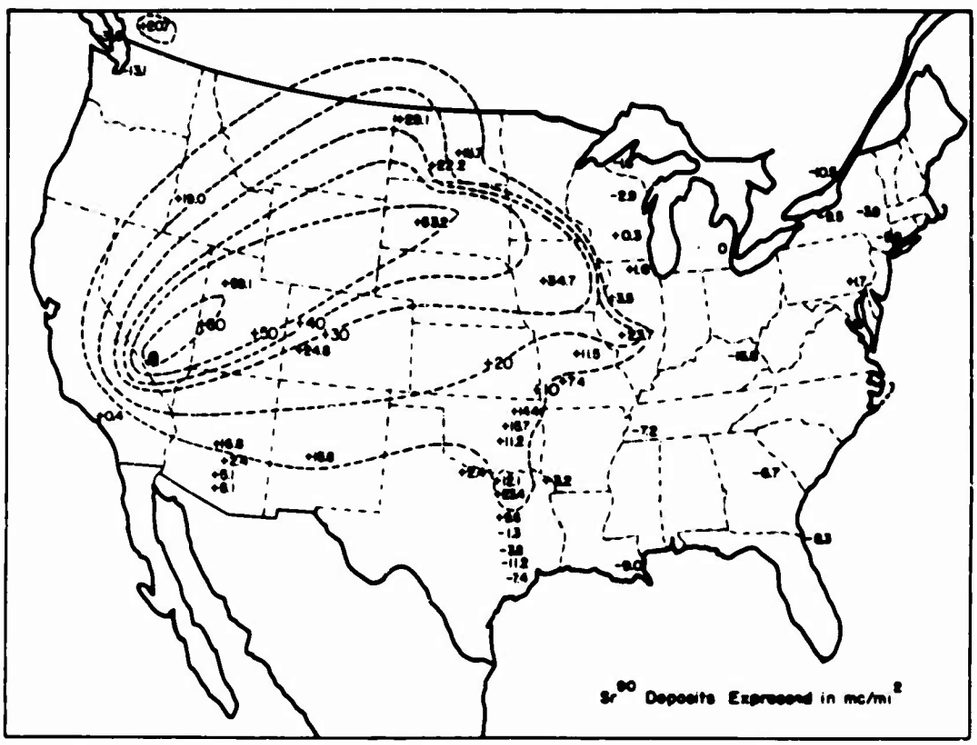


FIGURE 60 STRONTIUM-90 IN U.S. SOILS FROM INTERMEDIATE FALLOUT

fallout within the test site. It can be seen that the "excess" east of the Mississippi River is about -10 mc/mi^2. This reflects the fact that precipitation in this area usually comes from maritime air from the Gulf of Mexico. Consequently, little Nevada fallout appears here, and the amount that does appear is proportionately somewhat depleted of strontium-90 when compared to the rest of the 30° N to 50° N latitude band.

HASP Soil Studies

From the point of view of the external radiological health hazard it is important to know the deposition and distribution in soils of the gamma emitting nuclides such as cesium-137. Such information has generally been inferred from the strontium-90 deposits and the theoretical production ratios of the gamma active nuclides to strontium-90 in the fission process. Although this approach is reasonably satisfactory when only the total amount of deposited radioactivity is required, it is obviously of little use when the vertical distribution of the gamma activity is the unknown factor. Such information cannot, of course, be deduced immediately from the vertical distribution of strontium-90 within soils because of the different chemical properties of strontium and the other nuclides. In the case of cesium-137 an activity production ratio of Cs^{137}/Sr^{90} of 1.8 can be assumed and thus the total cesium-137 deposit within the soils can be calculated by multiplication of the strontium-90 deposit by the above ratio. However, because of the greater adsorption of cesium-137 on clay minerals compared to strontium-90 it is expected that the cesium-137 will tend to be more concentrated in the upper layers of the soils than the strontium-90. The exact distribution of cesium-137 with depth in soils can only be determined, therefore, by direct radiochemical or gamma spectroscopic analyses.

One of the objectives of the HASP program has been to define at least some

of the factors responsible for the distribution of radioactivity in soils. Within New Jersey there exists a wide range of soil types covering areas of excellent and poor drainage, high and low permeability, high and low organic content, and of course, disturbed and undisturbed sites. The undisturbed sites are those which have been subject only to natural processes during the past 10 years or so since radioactive fallout from nuclear weapons testing became appreciable. For these reasons several soil samples were collected in New Jersey during the summer of 1960. In addition a few samples were collected in the state of Kansas, an area of contrasting major soil classification. Samples were collected in the above areas with a specially designed coring device which was basically a steel cylinder with 1/4 inch wall thickness, two feet long and one foot in diameter. The soils were then returned to the laboratory where they were removed from the corer by an extrusion process, crushed, ground, and blended. Aliquots from several depths in the core were then separated for radiochemical analysis.

Concentrations of strontium-90, ruthenium-106, cesium-137 and cerium-144 in New Jersey soils were determined as a function of depth in the soil cores. For all four nuclides there was a sharp decrease in concentration with depth. In general the concentrations dropped by more than one order of magnitude in the first 6000 to 7000 grams of soil, which is equivalent to a depth in most soils of about 6 inches. The study showed that, as expected, strontium-90 penetrates to further depths in the soil and is more mobile than cesium-137. Direct comparison of these two nuclides with the remaining two nuclides, ruthenium-106 and cerium-144, is complicated by the fact that the latter two possess much shorter radioactive half-lives (~1 year) than the former pair (~30 years). It is difficult to correct the observed concentrations of the short-lived nuclides at a given depth in soil because the age of the material at any point cannot be assessed accurately. However, because of the

closeness of the half-lives of cerium-144 (285 days) and ruthenium-106 (1 year) some information on the movement of activity can be obtained from a comparison of the behavior of the two nuclides. The relative profiles of ruthenium-106 and cerium-144 indicate higher concentrations of cerium-144 at the top of the soil core and a more rapid decrease in concentration than ruthenium-106 for the first 1000 g. Beyond this depth, however, it is noticeable that the ruthenium-106 concentrations continue to fall quite rapidly whereas the cerium-144 results tend to show a diminished rate of decrease.

Table XX gives the average deposits of the four ratioactive nuclides strontium-90, ruthenium-106, cesium-137 and cerium-144 in New Jersey soils. The depth at which 100 percent of the total amount of fallout radioactivity was assumed to be present was 14,000 grams of dry soil. With the possible exception of cerium-144, it appears from this table, that the assumption of 100 percent of the activity being present in the first 14,000 grams of dry soil (~12 inches) is justified. The table also shows that the ratio of cesium-137/strontium-90 in the cumulative deposits to a depth of 14,000 g (~12 inches) is 2.1 $\pm$ 0.8 which is in accord with an initial activity ratio of 1.82 for the fast fission of uranium-238 and a value of 1.8 $\pm$ 0.5 determined from the analyses of air filter samples collected in the HASP program. Other analyses of soils for cesium-137 by Gustafson* have been performed. Combined with the strontium-90 results, which were determined by HASL, Gustafson obtained an average value for the cesium-137/strontium-90 ratio down to a depth of 6 inches of 1.62 $\pm$ 0.34. Also Gustafson recently reported that the cesium-137 concentration in soil near Argonne at the end of 1959 was approximately 200 mc/mi^2 compared to an average deposition in New Jersey of 155 mc/mi^2. The ratios of the cumulative deposits

*Gustafson, P. F., Science 130, 1404 (1959)

TABLE XX HASP SOIL DATA (NEW JERSEY)

Average Percentages of Total Activity in Soils as a Function of Depth

Nuclide	Depth (grams dry soil) 250	750	1500	2500	4000	6000	14,000
Strontium-90	16.7	34.1	49.6	63.6	76.4	85.1	100
Ruthenium-106	25.6	47.8	64.5	76.1	84.6	89.9	100
Cesium-137	28.7	55.3	71.7	80.9	88.2	93.4	100
Cerium-144	32.4	55.4	66.3	74.8	80.9	87.0	100

Average Vertical Profile of Strontium-90 in New Jersey Soils

Depth (inches)	Percentage of Cumulative Activity at 12 Inches Depth
1/2	15.4
1	31.3
2	54.5
3	68.8
4	78.7
6	88.4
9	95.6
12	100

Average Cesium-137/Strontium-90 Ratios with Depth in New Jersey Soils

Depth Increment (g dry soil)	Cs^{137}/Sr^{90} Ratio in Increment	Cumulative Cs^{137}/Sr^{90} Ratio
0-250	3.6	3.6
250-750	3.2	3.4
750-1500	2.2	3.0
1500-2500	1.4	2.6
2500-4000	1.2	2.4
4000-6000	1.2	2.3
6000-14000	0.9	2.1

Average Nuclide Ratios of Cumulative Activities in New Jersey Soils

Nuclides	Cumulative Activity Ratio (dpm/dpm at 14,000 g depth)
Cesium-137/Strontium-90	2.1 ± 0.8
Ruthenium-106/Strontium-90	2.3 ± 0.6
Cerium-144/Strontium-90	4.0 ± 1.0

of other nuclides to strontium-90 in New Jersey soils are also given in Table XX. Although these results agree quite well with predicted ratios, there are some large discrepancies between these results and those of Gustafson is soil analyses at Argonne. The results predicted from Gustafson's data are factors of 3.5 and 2.5 higher than these data for ruthenium-106 and cerium-144 respectively. A more complete discussion of this study is contained in Part III of the final HASP Report (DASA 1300).

In summary the following conclusions may be drawn regarding the deposition of strontium-90, ruthenium-106, cesium-137 and cerium-144 in New Jersey soils.

1. The average deposits of the above four nuclides in New Jersey were about 75,160,155 and 295 mc/mi^2 respectively on 1 July 1960. These values are in agreement with results for this area predicted on the basis of results of fission product determinations in precipitation and ground-level air.
2. Some discrepancies are observed between the concentrations of the short lived fission products ruthenium-106 and cerium-144 in the New Jersey and Chicago areas.
3. The average deviation of the cumulative deposits in New Jersey calculated from the results of analyses of 11 cores, is about 15 percent for strontium-90, ruthenium-106 and cerium-144. Cesium-137 results reflect a higher deviation of about 27 percent. With the exception of one soil the maximum and minimum cumulative activities did not differ by more than a factor of three of each other for all nuclides and all soils.
4. Vertical profiles of radioactivity within soils vary considerably and can be correlated with certain physical parameters. Permeability and drainage characteristics of the soil and underlying strata, appear to have profound influences on the distribution of radioactivity with depth.

5. In the "average" New Jersey soil in 1960 about 55 percent of the total deposit of strontium-90 is contained in the top 2 inches, about 79 percent in the top 4 inches and about 96 percent in the top 9 inches. These values contrast quite sharply with similar data observed by Schulert et al for soils collected in the same general area in 1958.

6. As expected strontium-90 appears to have penetrated to greater depths in the soils, on the average, than the other comparable long-lived nuclide cesium-137. Because of the difficulties involved in correcting the observed activities of cerium-144 and ruthenium-106 for radioactive decay the relative mobilities of the nuclides are somewhat obscured. Nevertheless, while ruthenium-106 shows relatively more penetration in the tops of the cores than cerium-144 the profile is unexplainably reversed at greater depths.

7. From the radiological hazard point of view the combined activities of the three gamma-emitting nuclides yield a dose rate on 1 July 1960 which is less than 3 percent of the external dose rate recommended by the NCRP for the general populace. This level of activity will, of course, decrease rather rapidly with time as the radioactivity decays (See Chapter XI).

CHAPTER XI

BIOLOGICAL HAZARD FROM FALLOUT

Introduction

The ultimate goal of all fallout studies is to assist in the assessment of the biological hazard from fallout, past, present or future. The HASP program has contributed considerable knowledge which may be used in this task. This includes: a measure of the amount of various radionuclides available for eventual deposition on the ground with subsequent biological exposure, rates of departure of stratospheric debris injected under varying meteorological and burst conditions, and eventual location of this material on the surface of the earth and in the biosphere.

In attempting to determine the effect of these fallout materials on man, a number of difficulties and uncertainties have arisen. As we have seen in previous chapters, there are uncertainties in the meteorological or physical aspects of distribution of the radionuclides upon the surface of the earth. However, the physical aspects can be said to be "well known" when compared to the problems inherent in determining such things as soil weathering, plant uptake, nuclide discrimination factors in the food chain, shielding from external exposure, calculation of effective dose rates to various organs, and so on. Perhaps the greatest uncertainty lies in the area of biological response as a function of varying types and rates of exposure to the ionizing radiation from various nuclides (including disruptive recoil of elements such as carbon-14). The reason for this latter unknown is that there have not been, nor is it likely that there ever will be, any individual casualties clearly attributable to the low levels of radiation which world-wide fallout has produced to date. The only hope of uncovering dose-effect relationships for these levels lies in: extensive epidemiological statistical studies, extrapolations from

higher doses and dose rates, and extrapolations to man from lower animals and plants.

Factors Affecting Radiation Hazard

In order to evaluate the long-term residual radiation hazard from world-wide fallout, a number of complex and interrelated concepts must be dealt with. First the type of exposure which may occur must be considered. This might include exposure from externally or internally located elements giving radiation doses either to the whole body or specific organs or localized regions of the body. Second the type of response the body makes to whatever kind of exposure it may receive must be considered. This might include genetic effects or somatic effects such as disease or accelerated aging. Third the types of radioelements present in the fallout material which could produce some specific response must be considered. Such factors as the abundance, type of radiation emitted, chemical nature, and method of reaching the organ to be exposed must be dealt with. Finally some yardstick must be established to measure the amounts of exposure and amounts of response to that exposure. After these steps have been taken the relative importance of various radionuclides, as far as hazard is concerned, can be established.

While the amount of response the human body makes to the low levels of radiation encountered so far from fallout is only vaguely known, nature has provided a yardstick of sorts in the form of natural background radiation. Since mankind has developed in the presence of this somewhat fluctuating background which itself may have been a large contributing factor in the evolution of the species found on earth today, we might assume that doses which are small compared to this background will not generally be harmful. This is the kind of value judgment which must be made cautiously, however.

Exposures to radiation can be broken into two basic categories: those that originate from material taken into the body and those that originate from material outside the body. In either case exposure to the whole body considered as a single organ can result. Gamma radiation outside the body can easily penetrate any cell in the body and when absorbed can produce damage. Gamma rays emitted inside the body can do the same thing. Beta rays emitted by some elements that spread uniformly through the body can also produce a whole body dose. Some specific organs of the body such as the gonads, lens of the eye or bone marrow may be especially susceptable to radiation. Critical organ doses by materials which preferentially concentrate in these areas may be of special importance. Iodine is preferentially concentrated in the thyroid gland while strontium and calcium are preferentially concentrated in the bones.

Any biological effect produced by radiation depends on the absorption of energy from the radiation. For many years the roentgen (r) has been used as a measure of x-and gamma-ray absorption in body tissue. Conceptually, the roentgen is only a measure of the ability of x- or gamma-rays to produce ionization in air and not of the absorption of these rays in tissue. More recently the absorbed dose of any radiation has been defined as "the energy imparted to matter by ionizing particles per unit mass of irradiated material at the place of interest." The unit of absorbed dose is the rad. For most purposes, the number of roentgens can be considered to be numerically equal to the number of rads in soft tissue.

The same absorbed dose of different kinds of radiation does not, in general produce the same biological effect. The different kinds of radiation have a different relative biological effectiveness (RBE). It is well known that the RBE for a particular kind of radiation may be highly dependent upon such factors as the specific

biological effect under consideration, the tissue irradiation, the radiation dose, and the rate at which it is delivered. The RBE dose is equal numerically to the product of the dose in rads and an agreed conventional value of the relative biological effectiveness (RBE). The unit of RBE dose is the rem, considered to be that dose which is biologically equivalent to one roentgen of x or gamma radiation. For example, one rad of neutrons is conventionally considered to be equivalent to 10 roentgens of gamma radiation, and this equivalence is expressed by saying that the RBE dose is 10 rem. However, it has been found experimentally that the same RBE dose of different radiation sources in the bone does not always produce the same biological effect. A numerical factor called the relative damage factor is introduced to take care of this difference. Thus, in the case of bone, the biological effect is equal to the product of the RBE dose and the relative damage factor.

Most delayed effects, in man, are inferred from consideration of experimental knowledge in animals, from available epidemiological statistical observations, and from a limited number of medical case observations. Delayed effects herein considered are those effects observable at some time following exposure. The effects considered are: (1) genetic effects; (2) somatic effects, including the appearance of leukemia, skin changes, precancerous lesions, neoplasms, cataracts, changes in the life span, and effects on growth and development. The delayed effects produced by ionizing radiation in an individual are not unique to radiation and are for the most part indistinguishable from those pathological conditions which are normally present in the population and which may be induced by other causes.

Biologically Important Nuclides in Fallout

As a result of consideration of the above factors and with a knowledge of the characteristics of the products of nuclear detonations, it is possible to eliminate

a large number of nuclides as possible sources of hazard in world-wide fallout. These are elements with short half lives, low production, little affinity for incorporation into food chains or the body, or weak radiations. The few remaining nuclides that appear to account for the majority of the dose to humans may be conveniently grouped into two classes, those that produce whole body radiation and those that produce localized radiation. The major single source of whole body radiation appears to be from cesium-137 deposited externally on the ground and internally from ingestion through the food chain. The shorter lived elements of zirconium-95, ruthenium-103,-106, and cerium-141,-144 also contribute a significant amount to the external dose. All of the above nuclides emit gamma radiation. Beta radiation from carbon-14 also may contribute to the whole body dose. The major source of localized radiation is that which comes from strontium-90 which is deposited in the bones. Another important source of localized radiation is that to the thyroid gland from iodine-131,-133. Since iodine-131 has a half life of only eight days, past exposure to this nuclide has been primarily confined to tropospheric fallout. While the largest dose rates to a single organ (the thyroid) of people in the general population have probably resulted from iodine-131 in tropospheric fallout, it is felt that it does not represent as great a world-wide hazard as does strontium-90. This is because the doses have been transient in nature.

A number of sampling networks have been established for the purpose of evaluating the amounts of exposure the general population has been and will be subjected to from fallout. These include: measurements in air and rainwater and soil to evaluate the amount and rate of accumulation on the surface of the earth; measurements in foods such as milk, wheat products, vegetables, meat etc., and measurements in people directly. Comparisons between the soil, rain, and food measurements provide a

method for evaluating the mechanisms of incorporation into the food chain. Soil measurements also allow calculation of the external gamma dose to be made. Comparisons of concentrations in people with those in food, etc., provide information on discrimination factors and the effects of varying diets. Continuing reports of data collected in the various networks along with interpretive comments may be found in "Radiological Health Data" published monthly by the U. S. Public Health Service and the Health and Safety Laboratory (HASL) Reports published quarterly by the U. S. Atomic Energy Commission.

Measurement Standards

A great deal of effort has been expended in recent years to establish yardsticks which can be used to measure the amount of danger involved in exposure to the radiations from various isotopes. Maximum permissible concentrations of various isotopes in the environment have been suggested by the ICRP and NCRP. The term "maximum permissible concentration" is somewhat misleading because it may suggest to some that concentrations below MPC insure immunity from deleterious effect while concentrations greater than MPC will surely cause a deleterious effect. This is not the case and both organizations have been quick to point this out. Recently the Federal Radiation Council has taken steps to establish a set of Radiation Protection Guides for normal peacetime operations. Like the MPC's, they are based primarily on background radiation levels supplemented by the sparse data which exists on radio-biological, dose-effects relationships.

The natural background radiation has always existed and actually was greater in the past than it is today. It consists of two parts, that of terrestrial origin and that of extra-terrestrial origin. The latter, known as cosmic radiation is variable and increases with latitude and altitude. The cosmic radiation besides

giving direct doses also produces carbon-14 which is itself radioactive. The background radiation of terrestrial origin comes from those naturally radioactive elements found in rocks and minerals. Table XXI shows the average annual dose to various organs from the natural background (medical and fallout doses are excluded). Broadly speaking, the background dose to the body lies roughly between 100 and 150 millirem/year but may rise to several times that value in some areas. The Federal Radiation Council has established a Radiation Protection Guide of 170 millirem for yearly whole-body exposure of average population groups. This guide, or dose which should not be exceeded without careful consideration for the reasons for the exposure, is in essential agreement with the recommendations of the NCRP, ICRP, and the National Academy of Science population genetic dose of 5 rem in 30 years (exclusive of natural radiation and medical x-ray exposure).

TABLE XXI

Natural Background Radiation

(Millirem per Year)

SOURCE	SOFT TISSUE	LUNGS	BONE	BONE MARROW
Cosmic Rays				
Sea Level	30	30	30	30
5,000 feet	70	70	70	70
Terrestrial Radiations	50	50	50	50
Internal Emitters				
K-40	20	20	9	9
C-14	2	2	2	2
Radon	2	100-1000	2	2
Radium	--	--	40	--

Whole Body Dose (External Sources)

The whole body dose from fallout from past nuclear tests is composed of radiations from external and internal sources. The external exposure rates from gamma ray emitting nuclides on the ground have been reported by a number of investigators. Vennart [106] has calculated that the open field dose in the UK from mixed fission products reached a peak of about one-half the natural background in 1959. Vohra[107] calculated from rainfall measurements in India that the 9 month peak in fallout between October 1958 and June 1959 would produce an infinity external gamma radiation dose of about 60 millirem. The Scandinavian measurements[96] show an increase of about 20% of background during August 1958.

Gustafson[108] has reported extensive results from measurements of various nuclides in soil collected near Chicago. Unshielded (open field) measurements taken at the collection site in April 1959 showed a dose rate of 18 microrem per hour. Calculations based on radiochemical analysis of the soil and expected soil shielding showed that as much as 8 microrem per hour came from fallout material. As a check, the natural background was "calculated" to be 11 microrem per hour. The calculated total of 19 microrem per hour checks well with the total of 18 microrem per hour measured *in situ*. It is reasonable to expect that a shielding factor of 5 can be applied to the open field dose when calculating average population exposure as Gustafson has done. Weathering has apparently not been an important factor in reducing the exposure to accumulations a few years old.

A fairly simple calculation can be made using Gustafson's soil measurements of cesium-137 to obtain the external open field dose from cesium-137 and the shorter-lived elements. It is reasonable to assume that half the cesium-137 which fell near Chicago during the last decade was from U. S. tests with a mean residence time

of about 500 days and that half came from Soviet tests with a mean residence time of about 200 days. It may be further assumed that there is little fractionation between nuclides, that their radioactive half lives and productions are those given in Chapter VI, that the specific doses expressed in (microrem/hr)/(1000 mc/mi^2) are those given by Gustafson and that the ultimate concentration of cesium-137 on the ground will be 220 mc/mi^2. The calculation of the open field 30 year exposure dose near Chicago leads to the results shown in Table XXII.

TABLE XXII

30 Year Open Field Dose 1955-1985 at Argonne

Nuclide	Mean Life*	Production	Specific Dose	Total Dose*** US Debris	Total Dose*** Soviet Debris
Zr-95	94	4.7%	6.33	35	69
Cs-137	14,600	5.4%	2.56	47	49
Ru-106	530	2.7%	0.85	9	12
Ru-103	58	4.5%	2.10	7	15
Ce-144	420	3.3%	0.18	2	3
Ce-141	46	4.3%	0.22	0	1
			Total	100 +	149 = 249 mr
					= 50 mr (shielded)

* in days
** in (microrem/hr)/(1000 mc/mi^2)
*** in millirem

The fraction of any one nuclide that decays prior to deposition is given by the expression:

$$f = \tau_m/(\tau_r + \tau_m) \qquad (19)$$

where τ_m and τ_r are, respectively, the meteorological mean residence time and

the radiological mean life. It can be seen that the Soviet debris contributes about 50% more dose than the U. S. debris due to its shorter residence time. Assuming a shielding factor of 5, it is noted that the total external dose from cesium-137 in 30 years is about 20 millirem and that for the shorter lived elements it amounts to about 30 millirem. During 1959, cesium-137 contributed about 1 millirem while those of shorter life contributed about 20 millirem to the shielded external dose.

Since there is considerable variability in fallout deposition from place to place in any one latitude band, it is difficult to assign a single set of values for the representative population doses to be expected from various nuclides in stratospheric fallout. Chicago lies sufficiently far from the Nevada test site that it is not unduly biased by tropospheric fallout from this source and use of the cumulative cesium-137 levels in this vicinity as a base line may fairly well represent the mid-continental U. S. It is noted that levels of Cs-137 in Chicago milk have not differed markedly from the national average. The 220 mc/mi^2 peak concentration of cesium-137 assumed to exist in the Chicago area appears to be somewhat higher than the average peak value to be expected in the 30^o N to 50^o N latitude band. Figure 59 shows that the strontium-90 peak will be about 75 mc/mi^2 which suggests that the cesium-137 peak should be no more than about 150 mc/mi^2.* Use of the 220 mc/mi^2 figure may then represent a slight overestimate and is therefore conservative. On the other hand we have assumed that the isotopes with a half-life of less than two weeks have decayed completely prior to deposition. This, of course, is not the case, especially for tropospheric fallout.

Whole Body Dose (Internal Sources)

Cesium-137 is a source of internal gamma exposure since it can be incorporated

* cf. New Jersey Soil data, Chapter X

into food chains. It is chemically somewhat like potassium and consequently is distributed in the muscle tissue of the body. A number of studies have been made of the concentrations of cesium-137 in food. Walton[110] has reviewed these studies and finds that the source of about one-half the cesium-137 ingested in the U. S. comes from milk alone. In addition, he notes that there is a growing body of evidence that shows that past concentrations of cesium-137 in foods and consequently in people have been more dependent on the rate of fallout on plants used for human or dairy animal consumption than on the cumulative amounts in the soil. This apparently is due to the fact that larger quantities of cesium-137 are deposited on the leaves of plants than are incorporated into the leaves by uptake from the soil through the root systems of the plants. Burton[109] has shown that only about 20% of the strontium-90 in milk in the U. K. in 1958 came from the soil. A similar situation apparently prevails in the case of cesium-137.

Due to its similarity to potassium, measurements of cesium-137 in the biosphere are usually made in terms of micromicrocuries of cesium-137 per gram of potassium (the cesium unit or C. U*). The expected body burden in C. U. and the resultant internal dose to the general population can be calculated from diet concentrations. The equilibrium body burden in C. U. should be about 1.8 times the average level in the diet. The dose in millirem per year is numerically about one-fiftieth the body burden in C. U. The most direct way of measuring the cesium-137 body burden, however, is through the use of a whole body counter. Table XXIII shows average values of cesium units measured in milk and in individuals in the U. S. along with the calculated dose since 1956**

* also known as the moonshine unit

** cf. Baarli, J., et. al., Radiocaesium and Potassium in Norwegians, Nature, 191, 436 (1961)

Several features can be seen in this data. First there was a gradual increase from 1956 to 1958 as the soil accumulated more and more cesium under conditions of constant fallout rate. In 1959 the fallout rate doubled and the concentrations in milk and in people also increased markedly. During 1960 when the fallout rates were dropping rapidly, the milk concentrations started dropping but the concentrations in people lagged behind. This lag may be partially due to the 140 day biological half-life of the cesium and partially due to consuming stored food which was grown during a period of high fallout rate. By late 1960 the concentrations in people started to drop off rapidly. If the early 1961 figure represents the ultimate concentration due to removal of the fallout rate factor, then the 30 year cesium-137 internal dose will be about 20 millirem.

TABLE XXIII

Internal Doses from Cesium-137

Year	Milk*	People*	Dose**
1956	24	41	0.8
1957	49	44	0.9
1958	57	54	1.1
1959	74	83	1.7
Early 1960	38	80	1.6
Late 1960	~ 20	46	0.9
Early 1961	~ 10	32	0.6
			6 Millirem

* concentration in micromicrocuries of Cs-137/gram K (C.U.)
** dose in millirem per year.

Carbon-14 is a radioactiv isotope of carbon which occurs in nature due to

cosmic ray bombardment of nitrogen. It has a half-life of about 5760 years and emits a weak beta-ray. Since carbon is widely distributed in the body, it produces internal whole body radiation and accounts for about one per cent of the natural background radiation to the body (1.6 mr/yr). Carbon-14 is also produced by neutron bombardment of nitrogen in the air during the course of a nuclear detonation. Recent data shows that the concentration of carbon-14 in the air has increased 25 per cent (see Chapter VII); however, the amount which will ultimately remain in the air after mixing with the carbon reservoir of the oceans will be about 1% of that produced naturally. Walton[110] has calculated that bomb-produced carbon-14 will then contribute a whole body dose of about 6 millirem during the next 30 years and about 90 millirem during the next 8000 years. These totals show that considered over the next few generations, carbon-14 is considerably less hazardous than cesium-137; but, if the genetic impact on the next 200 generations is to be considered, it may be comparable to the total dose received from cesium-137 and the other external emitters during the next two generations.

Single Organ Dose

The most significant single isotope from fission products in world-wide fallout which can produce a single organ dose is strontium-90. Since it is chemically similar to calcium, it can be deposited in the bones where it has a long biological half life. Concentrations in the bones and diets of people are often expressed in micromicrocuries of Sr-90/gram of calcium. This is the strontium unit or S. U.* Like cesium-137, a major source of strontium-90 in the typical U. S. diet is milk. Unlike cesium-137, however, biological discrimination factors tend to reduce the S. U. levels in the plant to man food chain. During 1959 the average U. S. diet contained about 15 S. U., however it dropped to about 10 S. U. in 1960.

* also known as the sunshine unit

This reflects the dependence of food levels upon the rate of fallout factor described above.* Walton[110] has shown that the C. U./S. U. ratio in U. S. milk has remained at about 6 to 1. It has been estimated that a constant diet level of about 200 S. U. would produce a body burden of about 50 S. U. which in turn would give a bone dose of about 150 millirem per year. Kulp[111] has made an extensive study of the concentrations of strontium-90 in man. His study shows that the segment of the population with the greatest bone concentration is children who were one year old in 1959. He estimates that this group will reach an average peak concentration of less the 3 S. U. and then will gradually drop off to about 0.9 S. U. Older segments of the population will reach the 0.9 S. U. equilibrium level sooner and adults will approach this level from below since their levels today are about 0.5 S. U. Figure 61 shows these concentrations and the estimated future levels in the concentration for three age groups. Butler[112] has shown that measurements made in teeth and bones in the U. K. are comparable to those reported by Kulp. These results are shown in Figure 62. Calculations of the cumulative dose in the one year old age group for the coming years can be made from the projection shown in Figure 61. By 1985 a total dose of about 75 millirem will have been received and by the end of the normal life span it will be about 100 millirems. This can be compared with a Radiation Protection Guide of 150 millirem per year.

Besides strontium-90, two other isotopes which may produce significant single organ doses are iodine-131 and plutonium. As we have mentioned before, iodine exposure comes almost exclusively from tropospheric fallout. Measurements of iodine-131 in fallout have been scanty and the concentrations in people and the subsequent effect is not well documented. Even though some may have had thyroid dose rates higher than in any other organ, the exposure has been transient and the total dose low due to the

*See also Menzel, R. G., D. L. Myhre, and H. Roberts, Jr., Science 134, 559 (1961)

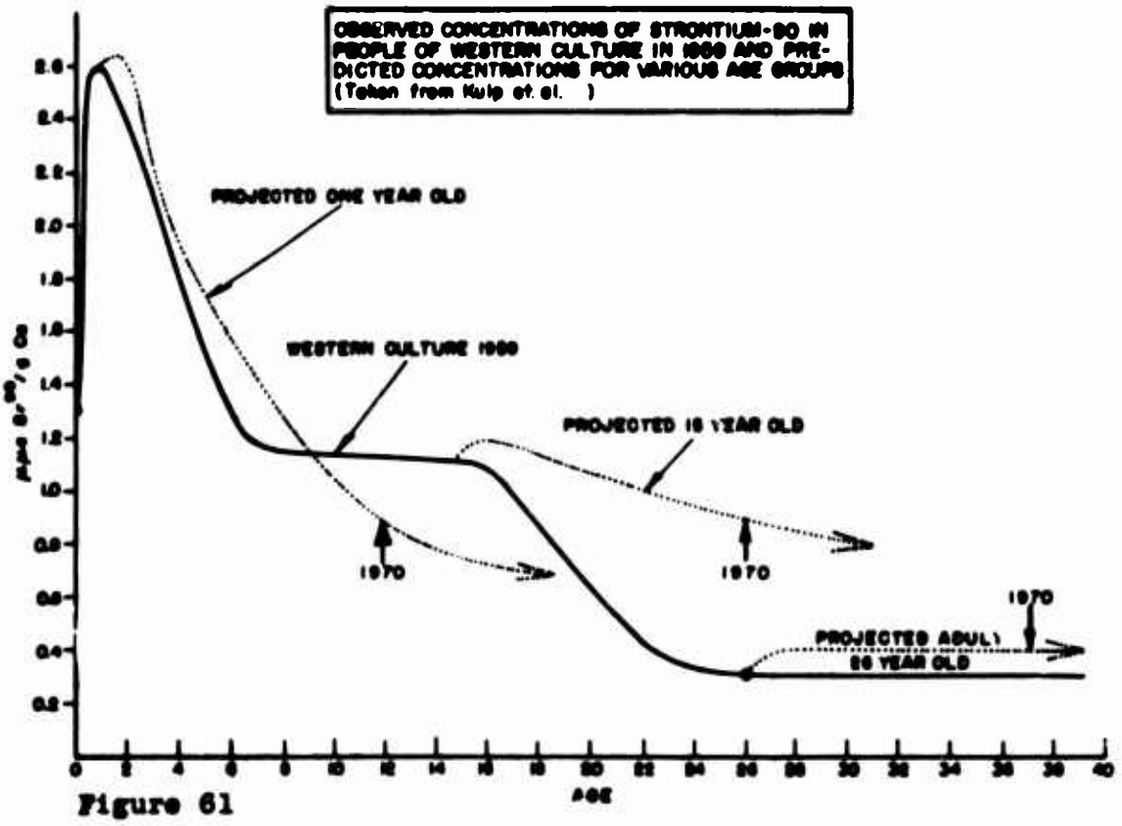


Figure 61

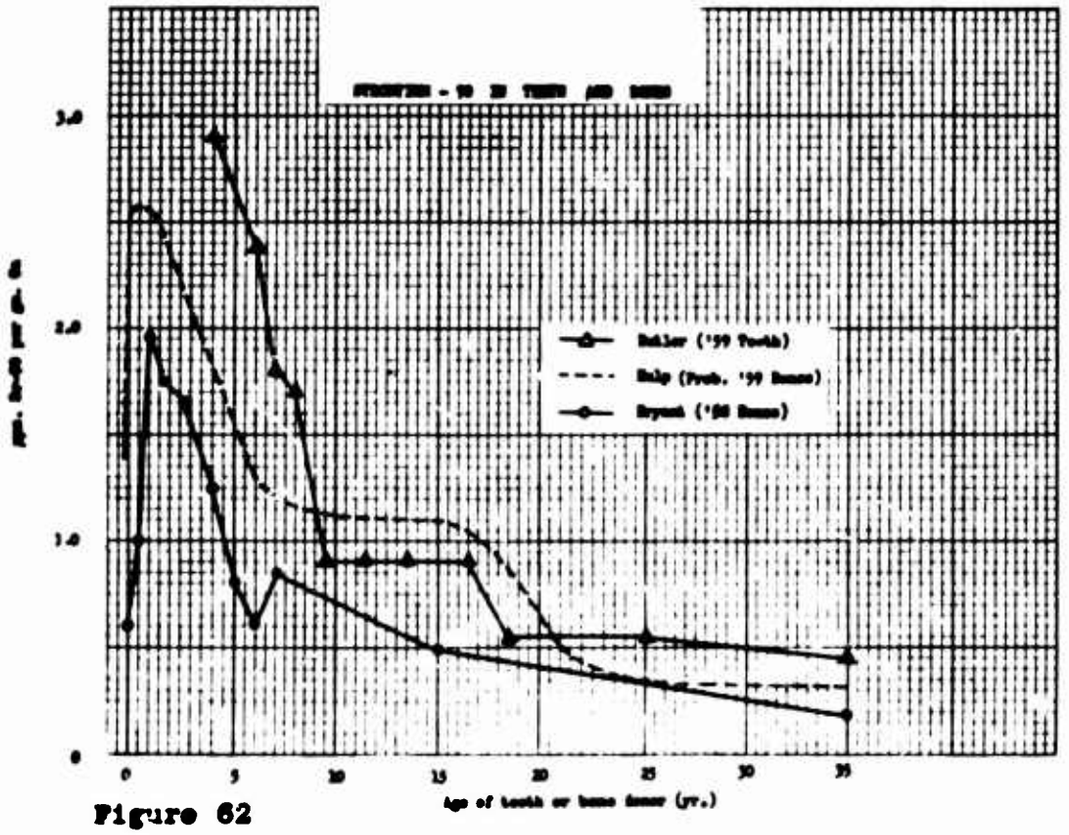


Figure 62

short half life of iodine-131. Moreover, there has been less total debris placed in tropospheric fallout than in stratospheric fallout, and the distribution of tropospheric fallout has not been as extensive as that of stratospheric fallout. For the above reasons it is felt that the hazard associated with iodine-131 is considerably less than that of strontium-90.

As part of the HASP program, several measurements of plutonium in human and animal tissue were made. Krey[113] reports that activity of plutonium in the pulmonary lymph nodes and gonads could be as high as ½% of the MPC. Inhalation is the suggested mode of entry and, if resuspension in dusty areas is effective, it may be that, in time, plutonium concentrations in the body could be comparable to those of strontium-90.

Summary of Past Tests

Table XXIV shows a summary of the radiation exposures from nuclear weapons tests. It is seen that during testing years, the annual exposure has reached as high as one-fourth the background levels of radiation. During the thirty year period from 1955 the whole body dose accumulated is about 2½% of the background. The 30 year whole body dose is considered to be the dose of genetic significance for any one generation. The lifetime dose from strontium-90 to the most susceptable element of the population (children born in 1958) is seen to be less than ½% of the RPG. If the whole body dose is added to this amount, the bone dose remains less than 1% of the RPG.* It is evident that the total effective dose from world-wide fallout is quite small even when compared to the fluctuations in the natural background exposure from organ to organ within the body and from place to place on the ground. Figure 63 shows graphically that radiation exposure from world-wide fallout produced by past nuclear tests falls at the bottom of the radiation exposure spectrum.

*but may be about 2% of the RPG for bone marrow (170 millirem/year)

TABLE XXIV

RADIATION EXPOSURE FROM NUCLEAR WEAPONS TESTS

(in millirem)

Whole body dose from:	1959 (1 yr)	1955-1985 (30 yr)	1955-2025 (70 yr)
Internal C-14	0.4	5	8
Internal Cs-137	2	20	30
External Cs-137	1	20	30
External Zr, Ru, Ce	20	30	30
Total	23	75	98
Natural Background (cosmic rays, rocks, etc.)	100	3,000	7,000
Additional bone dose to children from:			
Internal Sr^{90}	7	75	103
Population RPG for Sr^{90} †	500	15,000*	35,000*

†Intake guidance provided by the FRC is based upon 1/3 of the RPG

* The RPG is not usually applied to periods greater than one year

FUTURE TESTS

The questions is often asked, "what exposure can be expected if weapons testing were to be resumed?" While it is impossible to predict the course weapons testing might take, it may be instructive to estimate the ultimate levels of exposure which might be expected if the testing pattern of the years 1953-1958 were to be repeated indefinitely. The most evident consequence of such a situation is that the exposure levels would not continue to rise indefinitely, rather, they would tend to level off at some equilibrium value. One reason for this is the fact that part of the exposure is proportional to the rate of fallout and consequently if one has a steady fallout rate one will have a steady exposure level. In other words, a portion of the radioactive material has only a

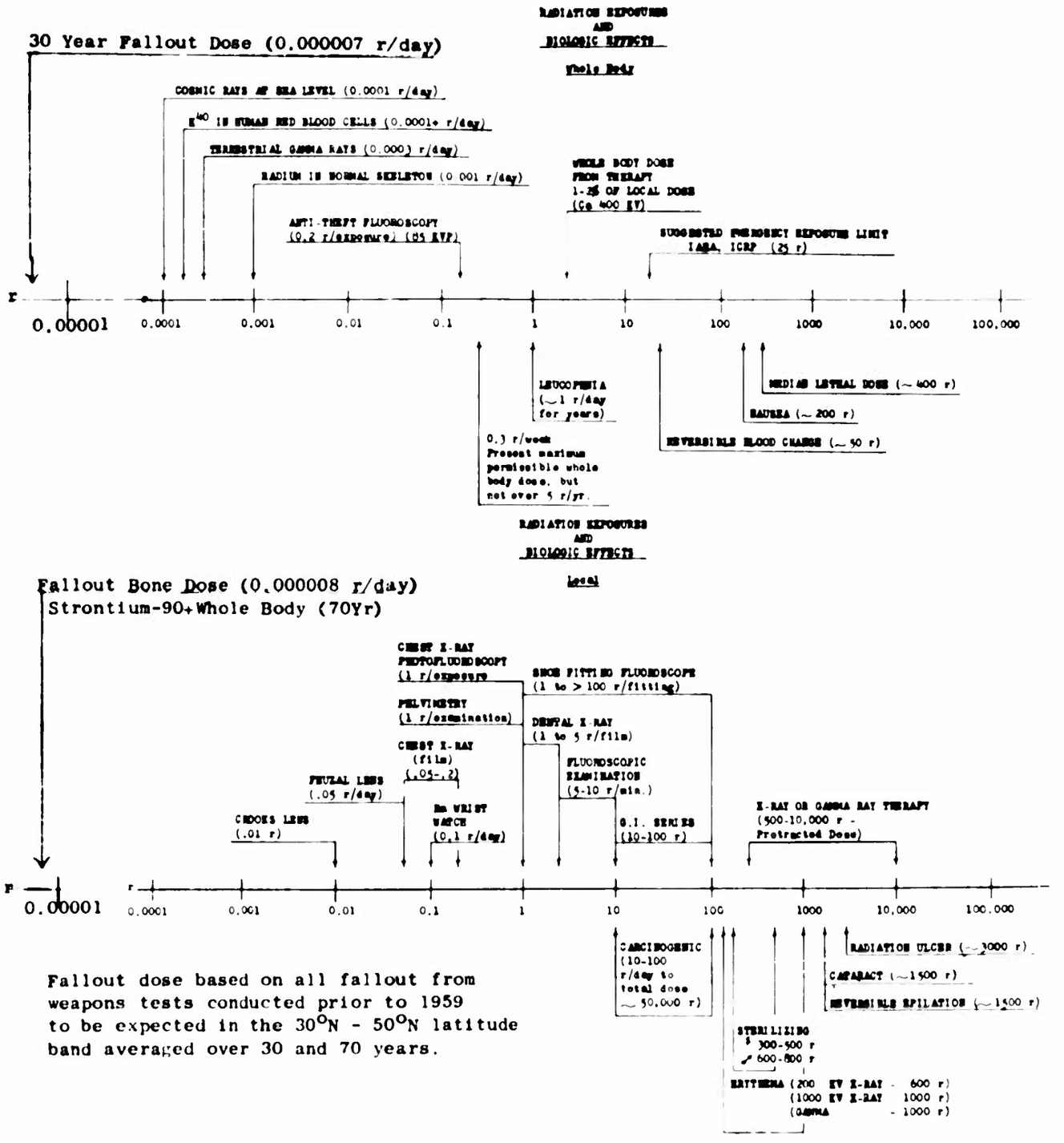

30 Year Fallout Dose (0.000007 r/day)
RADIATION EXPOSURES AND BIOLOGIC EFFECTS
Whole Body
COSMIC RAYS AT SEA LEVEL (0.0001 r/day)
K40 IN HUMAN RED BLOOD CELLS (0.0001+ r/day)
TERRESTRIAL GAMMA RAYS (0.0001 r/day)
RADIUM IN NORMAL SKELETON (0.001 r/day)
ANTI-THEFT FLUOROSCOPY (0.2 r/exposure) (65 KVP)
WHOLE BODY DOSE FROM THERAPY 1-2% OF LOCAL DOSE (Co 400 KV)
SUGGESTED OCCUPATIONAL EXPOSURE LIMIT IAEA, ICRP (25 r)
r
0.00001
0.0001
0.001
0.01
0.1
1
10
100
1000
10,000
100,000
LEUCOPENIA (~1 r/day for years)
MEDIAN LETHAL DOSE (~400 r)
NAUSEA (~200 r)
REVERSIBLE BLOOD CHANGES (~50 r)
0.3 r/week Present maximum permissible whole body dose, but not over 5 r/yr.
RADIATION EXPOSURES AND BIOLOGIC EFFECTS
Local
Fallout Bone Dose (0.000008 r/day)
Strontium-90+Whole Body (70Yr)
CHEST X-RAY FLUOROSCOPY (1 r/exposure)
SHOE FITTING FLUOROSCOPE (1 to > 100 r/fitting)
PELVIMETRY (1 r/examination)
DENTAL X-RAY (1 to 5 r/film)
CHEST X-RAY (film) (.05-.2)
FLUOROSCOPIC EXAMINATION (5-10 r/min.)
GONAD (.05 r/day)
Rn WRIST WATCH (0.1 r/day)
CROOKS LENS (.01 r)
G.I. SERIES (10-100 r)
X-RAY OR GAMMA RAY THERAPY (500-10,000 r - Protracted Dose)
F
r
0.00001
0.0001
0.001
0.01
0.1
1
10
100
1000
10,000
100,000
CARCINOGENIC (10-100 r/day to total dose ~50,000 r)
RADIATION ULCER (~3000 r)
CATARACT (~1500 r)
REVERSIBLE EPILATION (~1500 r)
STERILIZING ♀ 300-500 r ♂ 600-800 r
ERYTHEMA (200 KV X-RAY - 600 r) (1000 KV X-RAY 1000 r) (GAMMA - 1000 r)
Fallout dose based on all fallout from weapons tests conducted prior to 1959 to be expected in the 30°N - 50°N latitude band averaged over 30 and 70 years.

FIGURE 63

fleeting opportunity to produce exposure before it is effectively removed from the biosphere. Since the rate dependent exposure is associated with foliar deposition and since plant harvesting responds to annual seasonal cycles, it is appropriate to consider a one year time span as the interval of importance in calculating the rate effect.

Another characteristic of fallout that prevents exposure levels from rising indefinitely under continued testing is that of radioactive decay. Some shorter lived elements will have decayed almost completely by the end of one year. Others such as strontium-90 and cesium-137 will increase in quantity until their ultimate accumulation is about 40 times their average annual production. Carbon-14 would have an equilibrium inventory equal to about 8000 times the average annual production. Since the bulk of the past tests occurred during a 5 year period, the ultimate annual exposure rate from accumulated strontium-90 and cesium-137 would be about 8 times that due to the accumulated fraction of these elements from past tests. For carbon-14 it would be 1600 times, although the equilibrium point would not be approached for centuries.

Table XXV shows a rough estimate of expected equilibrium exposure levels expressed in millirem per year that might be expected under indefinite testing similar in character and amount to that conducted in the past. The figures in the table actually represent an upper limit to the exposure since they are based on the 1959 exposure levels shown in Table XXIV. The fallout rate in 1959 was greater than at any other time in the past, so the results given here are somewhat conservative since they overestimate the exposure to some extent. The 1959 exposure is divided into fractions due to accumulation and rate. For internal strontium-90 and cesium-137 about two-thirds is attributed to rate. For carbon-14 the exposure is all attributed to

TABLE XXV

ANNUAL RADIATION EXPOSURE*

	PAST TESTS (1959)		CONTINUED TESTS		
	Rate	Accumulation	Rate	Accumulation	
Internal C-14	½	.016	5	(25)**	
Internal Cs-137	1	½	1	4	
External Cs-137	0	1	0	8	
External Zr, Ru, Ce	18	2	18	3	
Whole Body Total			24	+ 15	= 39
Internal Sr-90	4	2½	4	+ 20	= 24

* in millirem/year
** not included in total

rate since only a small fraction of the carbon-14 has been incorporated into the large oceanic reservoir. Assuming a mean uptake residence time into this reservoir of about 50 years, the carbon-14 rate exposure could increase to 5 mr/yr. The eventual accumulation exposure due to carbon-14 could reach as high as 25 mr/year but it would take many thousands of years to do so. However, within the next century, this factor would be less than 1 mr/year, consequently, it is not included in the total. External Cs-137 exposure is considered to be mainly due to accumulation. The shorter lived elements are considered mainly rate dependent here because their half lives are short compared to the yearly time interval used. Only Ru-106 and Ce-144 have mean lives greater than one year. These two elements would therefore contribute a small fraction of the exposure in the succeeding year.

It can be seen from the table that the ultimate whole body dose could reach an upper limit of about 40 millirem per year which compares with the Radiation Protection Guide of 170 millirem per year. An additional dose to the bones of 25 millirem per year could be expected from strontium-90. This may be compared with the strontium-90 Radiation Protection Guide of 500 millirem per year. In general it is concluded that continued testing at the past rate would cause equilibrium exposure levels to be about three times the 1959 level. If testing were to be conducted for many thousands of years, additional carbon-14 accumulation could raise the equilibrium exposure level to four times that of 1959. In any case, the added amount would be considerably less than the present natural background radiation exposure levels.

It should be pointed out that the above results would be altered if the rate or character of testing were changed. If the rate were doubled, the exposure would double. If low atmospheric explosions were changed to upper atmosphere or near outer space explosions, the longer residence time in the mesophere would eliminate

exposure from the short lived elements but would reduce the exposure from strontium-90, cesium-137 and carbon-14 no more than 50%. If low air bursts were changed to surface bursts in remote regions much of the world-wide fallout material would be scavenged into local fallout to be deposited outside of the ecological reaches of mankind. Deep underground testing could further reduce the exposure by a substantial amount.

Chapter XII

CONCLUSIONS AND FUTURE WORK

General

The High Altitude Sampling Program has, to date, provided the most detailed and extensive study of radioactive material in the stratosphere carried on anywhere in the world. Over 100 million standard cubic feet of stratospheric air have been sampled. The program has fulfilled the task it set out to do, namely, to determine the role played by the stratosphere in the world-wide distribution of radioactive debris from low atmospheric weapons testing. The basic concept of the meridional network has been shown to be sound. Sampling in this network has provided inventories and distributions of material which help explain the major features of surface fallout noted by other programs both here and abroad. The U-2 can sample a major fraction of the stratosphere and good extrapolation may be made from balloon data up to 100,000 feet. Considerable information has been gained on the fate of debris injected into the mesosphere above 150,000 feet.

Recalibration of Ducts

Simultaneous sampling with the nose and hatch ducts revealed an apparent discrepancy in the flow rates through those ducts. The 20% difference was reduced to a 10% difference and at the same time the absolute flow rates were changed in such a manner as to increase the total inventories previously calculated by 8 to 13%. No evidence has been forthcoming that would suggest that the assumed filter paper efficiency of 100% is substantially wrong. As a result it is concluded that, considering variations in radiochemical reproducibility, variations in paper weight, non-uniformity of debris within the stratosphere, seasonal movement of debris within the stratosphere, and lack of vertical and latitudinal coverage, the absolute HASP inventories are good to $\pm$ 20% and the relative values of various averaged measurements

such as Ce-144/Sr-90 ratios in any region at one time are good to ± 10%. The problem of accounting for the outstanding 10% difference of flow rates between nose and hatch samples remains unresolved. Inventories based on both samplers are calculated as if there were no differences between them.

Stratospheric Measurements

The stratospheric measurements of a number of radionuclides has produced a set of data which is remarkably consistent internally. These data allow a detailed picture of mixing and transfer motions within and departing from the stratosphere to be constructed. An eddy or turbulent diffusion model of mixing supplies a simple explanation of the major features of the motion. Vertical diffusion proceeds at a lesser rate than does latitudinal diffusion and both are subject to variations in magnitude at different locations and with changes in season. Large scale meridional circulations as a transport mechanism seem to be ruled out by the HASP data.

The effective residence time for debris injected at various places within the stratosphere is now fairly well documented as are the conditions leading to exit from the stratosphere. While the relative importance of the various mechanisms which have been suggested for stratospheric-tropospheric interchange have not been determined, stratospheric measurements are consistent with the temporal and latitudinal distribution noted on the ground.

Fallout From Teak and Orange

Perhaps the most important result of HASP has been the measurement of the content and rate of removal of weapon debris from the stratosphere. In time, however, the same values could have been determined by surface sampling. One result that the HASP program has provided that surface sampling alone probably could not have provided is the fact that debris from the mesosphere has a half residence

time of about 5 years and that seasonal efflux in the polar regions provides the main avenue of escape for this material. The most significant conclusion to be drawn from the Teak and Orange data is that placing debris from weapons tests in the very top of the atmosphere will not substantially reduce the population dose from this material. While the 5 year residence time is sufficiently long to eliminate the shorter lived radionuclides, which account for one-third to one-sixth of the population dose, it is not long enough to reduce the population dose from strontium-90 and cesium-137 by more than 20%. In fact, it can be shown rather conclusively that a weapon burst on a land surface in the Pacific Ocean under conditions similar to many of the HARDTACK shots will produce less significant world-wide fallout than the same weapon burst at any altitude short of deep outer space. The reason for this, of course, is that more debris can be scavenged into local fallout from a land surface burst than can be expected to decay during its sojourn in the upper atmosphere.

Fallout Dose to Man

While the ultimate measure of the population dose which fallout materials may be expected to produce can be provided only by measurements on the surface and in the biosphere, proper prediction of this dose depends upon a knowledge of the amounts of debris available for future fallout and the rates at which this will occur. HASP has provided a great deal of information on these latter two topics. Considerable confidence can be placed in the prediction that surface concentrations of strontium-90 and cesium-137 will begin to diminish by late 1961. Since concentrations of these materials in food depend on fallout rate as well as total accumulation, the reduction in fallout rate in late 1959 engendered a concomitant reduction of concentrations of strontium-90 and cesium-137 in food and in people. However, due primarily to dietary habits there was a time lag of as much as a year

before this drop was reflected in the internal cesium-137 concentrations in people.

Finally, it is concluded that the expected population dose from world-wide fallout is insignificantly small. The 30 year genetically significant whole body dose will be less than 3% of the ubiquitous natural background radiation. The 70 year bone dose from strontium-90 and other radiations will be no more than 2% of the Radiation Protection Guide in that segment of the population most susceptible to the radiation, viz., children born in 1958. These percentages are smaller than the fluctuations in the natural background exposure from organ to organ within the body and from place to place on the globe.

Future Work

The HASP program, as such, concluded its sampling program in June 1960. The final report of this program (DASA 1300) will be completed by Isotopes, Incorporated by August 1961. Since June 1960 the HASP effort has merged with the cooperative DOD-AEC aircraft sampling program operated by the United States Air Force. Continued spot checks have been made in Australia (Crowflight Phases VI and VII) and elsewhere during November 1960 and May 1961. Limited sampling of stratospheric dust has been conducted on a continuous basis since June 1960.

Commencing in June 1961 limited HASP-like sampling was reactivated at Laughlin AFB, Texas. This program, under the nickname of STAR DUST, will extend the spot checks over a longer period of time in the mid-latitude regions. While the sampling will be more limited in extent than was the HASP program, its analytical effort will be expanded in scope. Its major goals are: to extend the HASP correlation with other sampling data, to elucidate further the fate of debris from Teak and Orange, and to provide a semi-empirical formulation for predicting fallout levels and population doses to be expected from the release of fission products in any region of the atmosphere as the result of the use of atomic energy in weapons or aerospace applications.

APPENDIX I

U-2 OPERATIONS

Considerable interest has been shown in the operational aspects of gathering the HASP samples with U-2 aircraft. It is therefore appropriate to provide here an illustrated description of a typical mission.

As is usual in the Air Force, the mission really starts a few days before take off. Extensive planning is necessary to insure the success of the mission. Since he must act as pilot, radio operator, navigator, flight engineer, and survival expert as well as scientific observer, each moment of the flight and each action the pilot must take is detailed in advance. Since celestial navigation is conducted, the sextant angles are precomputed based on forecast take-off time, winds aloft, expected time of observation and expected true air speed. Then lines of position are drawn on a strip map of the route on the basis of these precomputed angles. Corrections to these LOP's are also indicated if the sight is eventually taken a specified period of time early or late. In addition a composite navigation, communication and aircraft log is prepared along with the HASP flight data card. When Air Rescue Support is to be utilized in the mission, extensive coordination of emergency and communication procedures takes place.

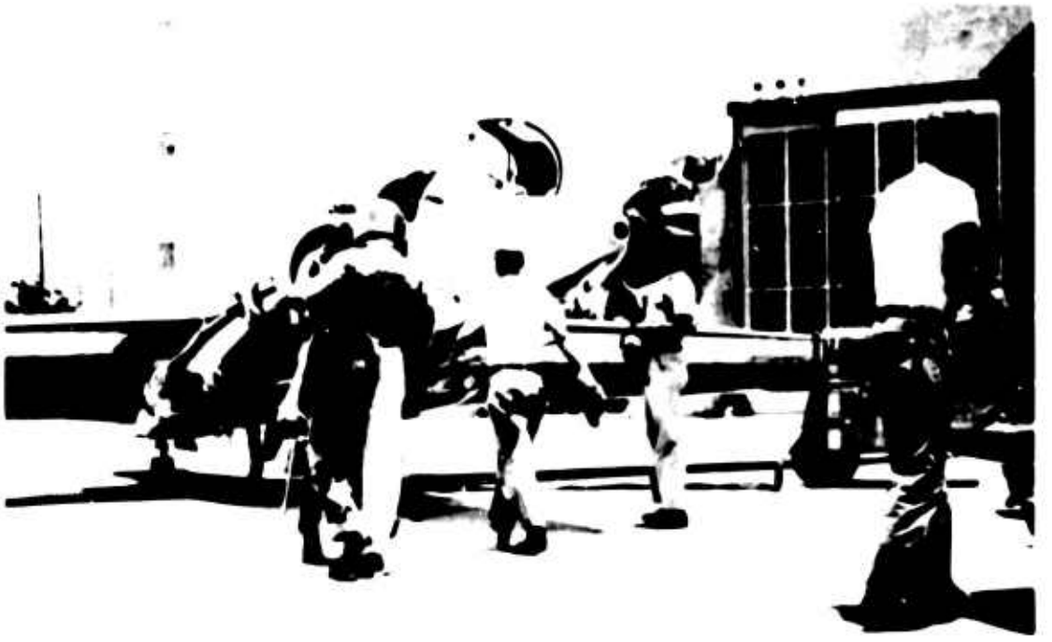
Several hours before the U-2 pilot is awakened for an early breakfast, the Air Rescue aircraft takes off and proceeds along the course. Since the U-2 flies about twice as fast as the Air Rescue aircraft, this procedure allows maximum coverage of the U-2's route. On the day of the mission the pilot must prebreathe oxygen

for several hours prior to take off. This procedure prevents decompression sickness if the U-2 cabin becomes depressurized during the flight. Considerable precautions are taken to insure that the pilots personal equipment and survival gear are in the proper condition. A partial pressure suit is worn in case of cabin depressurization since exposure to the ambient pressure at 70,000 feet would be fatal within minutes. Since the pilot is subjected to considerable physical stress during the prolonged flight at high altitude, a careful examination is made by the Flight Surgeon prior to take off and after landing. In the meantime the filter papers are inserted in their holders and installed in the changer. This mechanism then receives a last minute operational check.

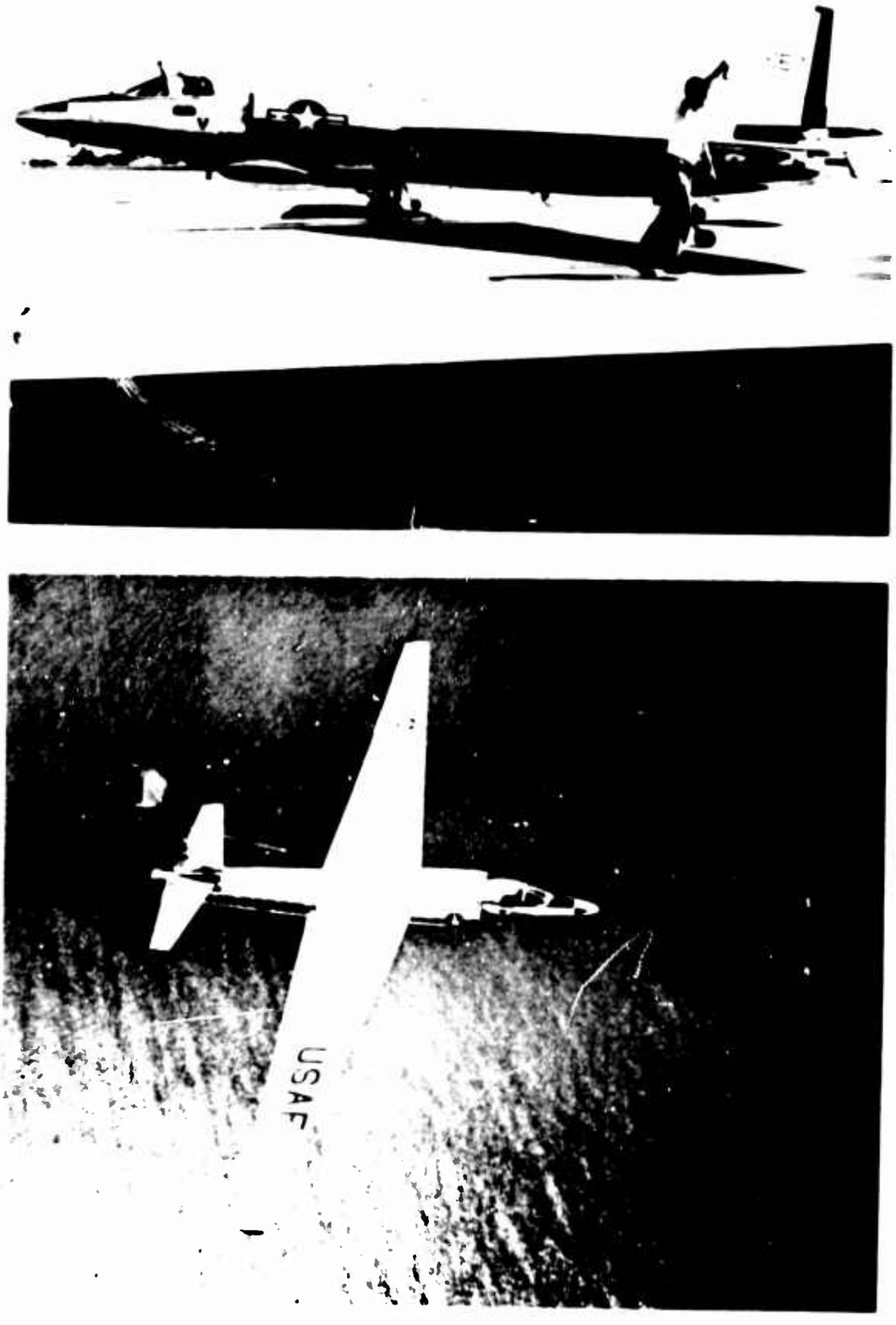
When the flight is ready to go, the pilot is assisted aboard. His personal equipment is rechecked and the aircraft is inspected by another pilot. A portable oxygen bottle is used while boarding the aircraft to en. e the efficacy of the prebreathing.

At the appointed time the pilot starts his engine and proceeds to the runway. After aligning the aircraft for take off, a ground crewman removes the safety pins from the outrigger gear which keep the wing tips off the ground. During the take-off roll the wings flex upward and the outriggers fall off. The pilot then climbs abruptly to the sampling altitude.

While flying the sampling route the pilot maintains his logs and flight data card and periodically changes the filter papers. He also sends position reports to the ground stations and maintains radio contact with the Air Rescue aircraft. The Air Rescue aircraft has a para-medical team to assist the pilot if he goes down. In addition this aircraft provides normal communications and navigational assistance as well as emergency assistance.



At the end of the route the U-2 pilot reverses course and changes altitude to sample inbound. The Air Rescue aircraft usually reverses course at about the same time. At the end of the flight the aircraft is stopped with the aid of a drag chute and the outriggers are reinstalled prior to abandoning the runway. Wing tip skids protect the aircraft if the wings tip down before it stops. A complete debriefing of the pilot is accomplished and the exposed filter papers are removed and placed into mailing envelopes. The mission is over when the Air Rescue aircraft returns, usually a few hours after the U-2.


USAF

APPENDIX II

CONVENIENT CONVERSION FACTORS AND CONSTANTS

Atmosphere

Mass of Mesosphere	4.32×10^{15} Kg = 1.24×10^{14}	1000 scf
Mass of Stratosphere	9.40×10^{17} Kg = 2.70×10^{16}	1000 scf
Mass of Atmosphere	5.26×10^{18} Kg = 1.51×10^{17}	1000 scf

Air Density at 273° K, 76 cm Hg 1.293 Kg/m^3 = 0.0807 Lb/ft^3

Air Density at 288° K, 76 cm Hg 1.226 Kg/m^3 = 0.0765 Lb/ft^3

1 Atmosphere = 1013 mb = 1034 cm H_2O = 76 cm Hg = 2116 psf

1000 scf = 34.7 Kg = 76.5 Lb = 28.3 m^3

Radioactivity

1 dpm/1000 scf = 28.8×10^{-3} dpm/Kg = 0.0354 dpm/m^3

1 dpm = 0.45 μ μcuries

Earth

Surface Area = 1.97×10^8 mi^2 = 5.06×10^{14} m^2

APPENDIX III

Summary of Nuclear Detonations

The following list of nuclear detonations has been compiled by Telegadas principally from press releases of the United States Atomic Energy Commission Public Information Service. However, reports in government publications and by government officials as to the size, type, cloud height, etc., of various explosions have been included when available. Press or unofficial estimates have not been included. Additional information is contained in HASL 111, April 1961.

The AEC has pointed out that this country does not disclose all of the USSR shots of which it has knowledge but limits itself to statements about explosions of special interest. The actual number of Soviet detonations is, therefore, higher than those announced. Similarly, not all of the detonations by this country have been announced. The United Kingdom and Republic of France detonations are understood to be the total number made by those countries.

The total fission yield in kilotons from testing through 1958 is given below:

YEAR	US & UK	USSR
1945	60	
1946	40	
1948	100	
1949 - 51	500	60
1952 - 54	37,000	500
1955 - 56	9,200	4,000
1957 - 58	19,000	21,000

Listed below are the approximate latitudes and longitudes of the various test sites:

U.S.

Nevada Test Site (NTS), U.S.A.	31°N 116°W
Eniwetok Proving Ground (EPG), Pacific	
Eniwetok	11°N 162°E
Bikini	11°N 165°E
Johnston Island, Pacific	17°N 169°W

U.K.

Monte Bello Islands, Australia	20°S 115°E
Woomera, Australia	31°S 137°E
Maralinga Proving Ground, Australia	30°S 131°E
Christmas Island, Pacific	2°N 157°W

U.S.S.R.

Arctic Test Site	75°N 55°E (Novaya Zemlya)
Siberian Test	52°N 78°E
U.S.S.R.	denotes the explosion was in Soviet territory, the test site was not given.

Republic of France

Reggan, Sahara Desert	27°N 0°

U.S. NUCLEAR DETONATIONS

No.	Date	Time (GMT)	Name	Weight of Burst (Ft.)	Type of Burst	Yield	- MSL: Feet - Cloud Top	Cloud Base	Trop	Location
	1945: OPERATION TRINITY									
1	16 July	1230	Trinity	100	Tower	19 KT	35,000			Alamagardo, N. M.
	WORLD WAR II									
1	5 Aug	2315		1850	Air Drop	Nominal				Hiroshima, Japan
2	9 Aug	0158		1850	Air Drop	Nominal				Nagasaki, Japan
	1946: OPERATION CROSSROADS									
1	30 June	2101	Able	518	Air Drop	Nominal	35,000			Bikini
2	24 July	2035	Baker	518	Underwater	Nominal	6,000			Bikini
	1948: OPERATION SANDSTONE									
1	14 Apr	1817	X-ray	200	Tower	37 KT	56,000	25,000	56,000	Eniwetok
2	30 Apr	1809	Yoke	200	Tower	49 KT	55,000	35,000	54,000	Eniwetok
3	14 May	1804	Zebra	200	Tower	18 KT	28,000	20,000	54,000	Eniwetok
	1951: OPERATION RANGER									
1	27 Jan	1345	Able	1060	Air Drop	1.3 KT	17,000		33,000	Nevada
2	28 Jan	1352	Baker	1060	Air Drop	7.8 KT	35,000		32,000	Nevada
3	1 Feb	1347	Easy	1080	Air Drop	1.0 KT	12,000		35,000	Nevada
4	2 Feb	1349	Baker-2	1100	Air Drop	8.9 KT	36,000		38,000	Nevada
5	6 Feb	1347	Fox	1435	Air Drop	22 KT	42,000		40,000	Nevada
	OPERATION GREENHOUSE									
1	7 Apr	1834	Dog	300	Tower				55,000	Eniwetok
2	20 Apr	1827	Easy	300	Tower	47 KT	40,000	30,000	54,000	Eniwetok
3	8 May	2130	George	200	Tower				55,000	Eniwetok
4	24 May	1817	Item	200	Tower				55,000	Eniwetok
	1951: OPERATION BUSTER-JANGLE									
1	22 Oct	1400	Able	100	Tower	< 0.1 KT	8,000	6,700		Nevada
2	28 Oct	1520	Baker	1118	Air Drop	3.5 KT	29,000	23,000	39,000	Nevada
3	30 Oct	1500	Charlie	1132	Air Drop	14 KT	40,000	32,000	38,000	Nevada
4	1 Nov	1530	Dog	1417	Air Drop	21 KT	40,000	27,000	38,000	Nevada
5	5 Nov	1630	Easy	1314	Air Drop	31 KT	45,000	31,000	35,000	Nevada
6	19 Nov	1700	Sugar	4	Surface	1.2 KT	16,000	11,000		Nevada
7	29 Nov	2000	Uncle	-17	Underground	1.2 KT	11,000			Nevada
	1952: OPERATION TUMBLER-SNAPPER									
1	1 Apr	1700	Able	700	Air Drop	1.1 KT	16,000		42,000	Nevada
2	15 Apr	1730	Baker	1109	Air Drop	1.2 KT	16,000	10,000	38,000	Nevada
3	22 Apr	1730	Charlie	944	Air Drop	31 KT	42,000	31,000	38,000	Nevada
4	1 May	1630	Dog	1040	Air Drop	19 KT	42,000	28,000	38,000	Nevada
5	7 May	1215	Easy	300	Tower	12 KT	34,000		41,000	Nevada
6	25 May	1200	Fox	300	Tower	11 KT	41,000		37,000	Nevada
7	1 June	1155	George	300	Tower	15 KT	37,000		37,000	Nevada
8	5 June	1155	How	300	Tower	14 KT	41,000		40,000	Nevada
	OPERATION IVY									
1	31 Oct	1915	Mike		Surface	Hydrogen Bomb	~100,000		56,000	Eniwetok
2	15 Nov	2330	King	1480	Air Drop	High Yield	~ 70,000		55,000	Eniwetok
	1953: OPERATION UPSHOT-KNOTHOLE									
1	17 Mar	1320	Annie	300	Tower	16 KT	41,000	28,000	36,000	Nevada
2	24 Mar	1310	Nancy	300	Tower	24 KT	42,000	26,000	40,000	Nevada
3	31 Mar	1300	Ruth	300	Tower	0.2 KT	14,000	11,000	36,000	Nevada
4	6 Apr	1530	Dixie	6920	Air Drop	11 KT	43,000	33,000	36,000	Nevada
5	11 Apr	1245	Ray	100	Tower	0.2 KT	13,000	8,000	38,000	Nevada
6	18 Apr	1235	Badger	300	Tower	23 KT	35,000	23,000	39,000	Nevada
7	25 Apr	1730	Simon	300	Tower	43 KT	45,000	31,000	38,000	Nevada
8	8 May	1530	Encore	2425	Air Drop	27 KT	41,000	29,000	39,000	Nevada
9	19 May	1205	Harry	300	Tower	32 KT	43,000	27,000	42,000	Nevada
10	25 May	1530	Grable	524	Gun	15 KT	38,000	23,000	38,000	Nevada
11	4 June	1115	Climax	1334	Air Drop	61 KT	43,000	35,000	39,000	Nevada
	1954: OPERATION CASTLE									
1	28 Feb	1845	Bravo		Surface	15 MT	114,000	55,000	55,000	Bikini
2	26 Mar	1830	Romeo		Barge				55,000	Bikini
3	6 Apr	1820	Koon		Surface	~ 100 KT			53,000	Bikini
4	25 Apr	1810	Union		Barge				57,000	Bikini
5	4 May	1810	Yankee		Barge				55,000	Bikini
6	13 May	1820	Nectar		Barge				56,000	Eniwetok
	1955: OPERATION TEAPOT									
1	18 Feb	2000	Wasp	762	Air Drop	1.2 KT	22,000	15,000		Nevada
2	22 Feb	1345	Moth	300	Tower	2.4 KT	25,000	16,000		Nevada
3	1 Mar	1330	Tesla	500	Tower	6.8 KT	30,000	18,000	38,000	Nevada
4	7 Mar	1320	Turk	500	Tower	43 KT	44,000	36,000	40,000	Nevada
5	12 Mar	1320	Hornet	300	Tower	3.6 KT	35,000	27,000	38,000	Nevada
6	22 Mar	1305	Bee	500	Tower	8.1 KT	40,000	29,000	37,000	Nevada
7	23 Mar	2030	Ess	-67	Underground	1.1 KT	12,000		39,000	Nevada
8	29 Mar	1255	Apple I	500	Tower	14 KT	32,000	22,000	39,000	Nevada
9	29 Mar	1800	Wasp Prime	740	Air Drop	3.2 KT	32,000		40,000	Nevada
10	6 Apr	1800	HA	30,820 MSL	Missile	3.3 KT	55,000		31,000	Nevada
11	9 Apr	1230	Post	300	Tower	1.5 KT	16,000	13,000		Nevada
12	15 Apr	1415	Met	400	Tower	22 KT	40,000	31,000	37,000	Nevada
13	5 May	1210	Apple-2	500	Tower	29 KT	43,000	34,000	41,000	Nevada
14	15 May	1200	Zucchini	500	Tower	28 KT	35,000	25,000	44,000	Nevada
	OPERATION WIGWAM									
1	14 May	2000	Wigwam	-2000	Underwater	30 KT				29°N 126°W
	1956: SAFETY									
1	18 Jan	2100			Surface	Slight				Nevada
	OPERATION REDWING									
1	4 May	1825	Lacrosse		Surface	Kiloton Range			53,000	Eniwetok
2	20 May	1751	Cherokee	4320	Air Drop	Several Megatons			53,000	Bikini
3	27 May	1756	Zuni		Surface				51,000	Bikini
4	30 May	1815	Erie	300	Tower				54,000	Eniwetok
5	6 June	0055	Seminole		Surface				52,000	Eniwetok
6	11 June	1826	Flathead		Barge				50,000	Bikini
7	11 June	1826	Blackfoot	200	Tower				52,000	Eniwetok
8	16 June	0114	Osage	680	Air Drop				52,000	Eniwetok
9	25 June	1806	Dakota		Barge				54,000	Bikini
10	8 July	1806	Apache		Barge				52,000	Eniwetok
11	10 July	1756	Navajo		Barge				50,000	Bikini
12	20 July	1746	Tewa		Barge				52,000	Bikini
13	21 July	1816	Huron		Barge				51,000	Eniwetok

U.S. NUCLEAR DETONATIONS

No.	Date	Time (GCT)	Name	Height of Burst (ft.)	Type of Burst	Yield	- MSL: Feet - Cloud Top	Cloud Base	Trop	Location
	1957: OPERATION PLUMBBOB									
1	28 May	1155	Boltzmann	500	Tower	12 KT	33,000	23,000	41,000	Nevada
2	2 June	1155	Franklin	300	Tower	140 Tons	17,000	14,000		Nevada
3	5 June	1145	Lassen	500	Balloon	0.5 Tons	7,000		43,000	Nevada
4	18 June	1145	Wilson	500	Balloon	10 KT	35,000	25,000	40,000	Nevada
5	24 June	1330	Priscilla	700	Balloon	37 KT	43,000	24,000	49,000	Nevada
6	5 July	1140	Hood	1500	Balloon	74 KT	48,000	35,000	53,000	Nevada
7	15 July	1130	Diablo	500	Tower	17 KT	32,000	20,000	43,000	Nevada
8	19 July	1400	John	20,000 MSL	Rocket	~ 2 KT	44,000		48,000	Nevada
9	24 July	1150	Kepler	500	Tower	10 KT	28,000	20,000	34,000	Nevada
10	25 July	1330	Owen	500	Balloon	9.7 KT	35,000	20,000	49,000	Nevada
11	26 July	0800	Pascal A		Underground	Slight	6,000			Nevada
12	7 Aug	1225	Stokes	1500	Balloon	19 KT	37,000	27,000	47,000	Nevada
13	18 Aug	1200	Shasta	500	Tower	17 KT	32,000	16,000	50,000	Nevada
14	23 Aug	1230	Doppler	1500	Balloon	11 KT	38,000	23,000	43,000	Nevada
15	30 Aug	1240	Franklin Prime	750	Balloon	4.7 KT	32,000	21,000	32,000	Nevada
16	31 Aug	1230	Smoky	700	Tower	44 KT	38,000		35,000	Nevada
17	2 Sept	1240	Galileo	500	Tower	11 KT	37,000	17,000	39,000	Nevada
18	6 Sept	1245	Wheeler	500	Balloon	197 Tons	17,000	14,000	50,000	Nevada
19	6 Sept	2005	Coulomb B		Surface	0.3 KT	18,000		50,000	Nevada
20	8 Sept	1300	La Place	750	Balloon	1 KT	20,000	14,000	44,000	Nevada
21	14 Sept	1645	Fizeau	500	Tower	11 KT	40,000	27,000	43,000	Nevada
22	16 Sept	1250	Newton	1500	Balloon	12 KT	32,000	19,000	52,000	Nevada
23	19 Sept	1700	Rainier	-790	Underground	1.7 KT	0			Nevada
24	23 Sept	1230	Whitney	500	Tower	19 KT	30,000	18,000	53,000	Nevada
25	28 Sept	1300	Charleston	1500	Balloon	12 KT	32,000	20,000	45,000	Nevada
26	7 Oct	1300	Morgan	500	Balloon	8 KT	40,000	26,000	37,000	Nevada
	1957: SAFETY									
1	6 Dec	2015			Vertical Shaft	Slight				Nevada
2	9 Dec	2000			Surface	Slight				Nevada
	1958: OPERATION HARDTACK - PHASE I									
1	28 Apr	0240	Yucca	86,000	Balloon				53,000	14° 37' N 165° 01' E
2	5 May	1815	Cactus		Surface				51,000	Eniwetok
3	11 May	1750	Fir		Barge				54,000	Bikini
4	11 May	1815	Butternut		Barge				53,000	Eniwetok
5	12 May	1830	Koa		Surface				57,000	Eniwetok
6	16 May	0130	Wahoo	-500	Underwater				59,000	Eniwetok
7	20 May	1830	Holly		Barge				52,000	Eniwetok
8	21 May	2120	Nutmeg		Barge				54,000	Bikini
9	26 May	0200	Yellowwood		Barge				55,000	Eniwetok
10	26 May	1800	Magnolia		Barge				54,000	Eniwetok
11	30 May	0215	Tobacco		Barge				55,000	Eniwetok
12	31 May	0300	Sycamore		Barge				55,000	Bikini
13	2 June	1845	Rose		Barge				57,000	Eniwetok
14	8 June	2315	Umbrella	-150	Underwater				54,000	Eniwetok
15	10 June	1730	Maple		Barge				53,000	Bikini
16	14 June	1730	Aspen		Barge				52,000	Bikini
17	14 June	1830	Walnut		Barge				54,000	Eniwetok
18	18 June	0300	Linden		Barge				54,000	Eniwetok
19	27 June	1730	Redwood		Barge				52,000	Bikini
20	27 June	1830	Elder		Barge				52,000	Eniwetok
21	28 June	1930	Oak		Barge				50,000	Eniwetok
22	29 June	0000	Hickory		Barge				51,000	Bikini
23	1 July	1830	Sequoia		Barge				52,000	Eniwetok
24	2 July	1730	Cedar		Barge				51,000	Bikini
25	5 July	1830	Dogwood		Barge				52,000	Eniwetok
26	12 July	0330	Poplar		Barge				55,000	Bikini
27	22 July	0420	Juniper		Barge				51,000	Bikini
28	22 July	2030	Oliver		Barge				48,000	Eniwetok
29	26 July	2030	Pine		Barge				52,000	Eniwetok
30	1 Aug	1050	Teak	252,000	Rocket	Megaton Device				Johnston Is.
31	12 Aug	1030	Orange	~100,000	Rocket	Megaton Device				Johnston Is.
	OPERATION HARDTACK - PHASE II									
1	12 Sept	2000	Otero	-400	Underground	38 Tons	9,000			Nevada
2	17 Sept	1730	Bernalillo	-456	Underground	15 T	7,500	5,500		Nevada
3	19 Sept	1400	Eddy	500	Balloon	83 T	11,000	7,500	48,000	Nevada
4	21 Sept	1500	Luna	-494	Underground	1.5 T	Low diffuse cloud			Nevada
5	26 Sept	2000	Valencia	-404	Underground	3 T	5,500			Nevada
6	28 Sept	0000	Mars		Underground	13 T	Low diffuse cloud			Nevada
7	29 Sept	1405	Nora	1500	Balloon	2 KT	18,500	10,000	40,000	Nevada
8	5 Oct	1410	Hidalgo	377	Balloon	77 T	12,000	8,000		Nevada
9	5 Oct	1615	Colfax	-350	Underground	5.5 T	5,500	4,500		Nevada
10	6 Oct	2200	Tamalpais	-330	Underground	72 T	Low diffuse cloud			Nevada
11	10 Oct	1430	Quay	100	Tower	7? T	10,000	7,500		Nevada
12	13 Oct	1320	Lea	1500	Balloon	1.4 KT	17,000	12,000		Nevada
13	14 Oct	1800	Neptune	-496.5	Underground	115 T	11,000			Nevada
14	15 Oct	1600	Hamilton	50	Tower	1.2 T	6,000	4,500		Nevada
15	16 Oct	0600	Logan	-830	Underground	5 KT	0			Nevada
16	16 Oct	1420	Dona Ana	450	Balloon	37 T	11,000	6,500	49,000	Nevada
17	17 Oct	2300	Vesta		Surface	24 T	10,000			Nevada
18	18 Oct	1425	Rio Arriba	72.5	Tower	80 T	13,500	11,000		Nevada
19	22 Oct	1330	Socorro	1450	Balloon	6 KT	20,000	20,000		Nevada
20	22 Oct	1650	Wrangell	1500	Balloon	115 Tons	10,000	7,000		Nevada
21	22 Oct	2340	Rushmore	500	Balloon	188 Tons	11,500		42,000	Nevada
22	24 Oct	1500	Catron	72.5	Tower	21 Tons	8,500	5,000		Nevada
23	24 Oct	1601	Juno		Surface	1.7 Tons	5,500			Nevada
24	26 Oct	0400	Ceres	25	Tower	0.7 Tons	6,000			Nevada
25	26 Oct	1020	Sanford	1500	Balloon	4.9 KT	26,000	12,500		Nevada
26	26 Oct	1600	De Baca	1500	Balloon	2.2 KT	17,500	10,000		Nevada
27	27 Oct	1430	Chavez	52.5	Tower	0.6 Tons	6,500			Nevada
28	29 Oct	0000	Evans	-848	Underground	55 Tons	0			Nevada
29	29 Oct	1445	Humboldt	25	Tower	7.8 Tons	7,500	6,000		Nevada
30	30 Oct	0300	Santa Fe	1500	Balloon	1.3 KT	18,000	13,000	39,000	Nevada
31	30 Oct	1500	Blanca	-835	Underground	19 KT	7,700			Nevada
32	30 Oct	2030	Titania	25	Tower	0.2 Tons	6,000			Nevada
	OPERATION ARGUS									
1	27 Aug		Argus-1	~ 300 miles	Rocket	1-2 KT				South Atlantic
2	30 Aug		Argus-2	~ 300 miles	Rocket	1-2 KT				South Atlantic
3	6 Sept		Argus-3	~ 300 miles	Rocket	1-2 KT				South Atlantic

U.S.S.R. NUCLEAR DETONATIONS

No.	Date	Type	Yield	Location	Remarks
1	[illegible] 29 Aug			USSR	First Russian nuclear detonation.
1	1/24 5 Oct*			USSR	Statement by Press Secretary Short, detonated recently.
2	22 Oct*			USSR	Statement by White House.
1	1/31 12 Aug		Thermonuclear	USSR	Part of a series.
2	23 Aug		Fission	USSR	Part of a series, energy release equivalent to type detonated at Nevada Test Site.
1	1/56 26 Oct*			USSR	Part of a series, statement by Mr. Strauss, series began in mid-Sept. and continued at intervals to the present.
1	1/33 4 Aug*			USSR	Part of a series statement by Dr. Libby - detonated in the past few days.
2	26 Sept*			USSR	Part of a series, statement by Mr. Strauss - detonated recently.
3	10 Nov*			USSR	Part of a series, statement by Mr. Strauss - detonated recently.
4	23 Nov*	Air Drop	Megaton Range	USSR	Part of a series, statement by Mr. Strauss.
1	1/56 21 Mar*			USSR	Part of a series, statement by Mr. Strauss - detonated in the past few days.
2	2 Apr*			USSR	Part of a series, statement by Mr. Strauss - detonated recently.
1	26 Aug		Large Type a Megaton	Siberia	Part of a series.
2	30 Aug		Large Test	Siberia	Part o. a series.
3	2 Sept			USSR	Part of a series
4	10 Sept			USSR	Part of a series, announced by Soviet Union.
5	17 Nov		Large Test	USSR	Part of a series.
1	1/57 1? Jan			USSR	Part of a series.
2	8 Mar			USSR	Part of a series.
3	3 Apr			USSR	Part of a series.
4	6 Apr			USSR	Part of a series.
5	13 Apr		Large	USSR	Part of a series.
6	12 Apr			USSR	Part of a series.
7	16 Apr		Large	Siberia	Part of a series-largest tested so far for this series.
1	1/57 28 Aug		Substantial Size	Siberia	
2	9 Sept*		Moderate Intensity	Siberia	Statement by Mr. Strauss, detonated within the past two days.
3	24 Sept		Megaton Range	Arctic	
4	6 Oct		Hydrogen Device	USSR	Announced by Soviet Union as a hydrogen device, AEC said it was of substantial size.
5	10 Oct		Small Explosion	Arctic	
6	28 Nov			Siberia	
1	1/58 23 Feb		Megaton Range	Arctic	
2	27 Feb		Megaton Range	Arctic	
3	27 Mar		Large	Arctic	
4	14 Mar		Not in Megaton Range	Arctic	
5	14 Mar		Not in Megaton Range	Siberia	
6	15 Mar		Below Megaton Range	Siberia	
7	20 Mar		Small Range	Arctic	
8	21 Mar			Siberia	This detonation was in a larger range than the test the day before.
9	22 Mar		Medium Range	Arctic	
1	30 Sept		Moderate to High	Arctic	
2	30 Sept		Moderate to High	Arctic	
3	8 Oct		Moderate	Arctic	
4	8 Oct		Moderate	Arctic	
5	5 Oct			Arctic	Lesser yield than the detonations (4) which occurred on 30 Sept. and 7 Oct.
6	10 Oct		Relatively Large	Arctic	
7	12 Oct		Largest	Arctic	
8	13 Oct		Largest	Arctic	
9	16 Oct		Largest	Arctic	
10	17 Oct		Small	Arctic	
11	20 Oct		Largest	Arctic	
12	22 Oct		Largest	Arctic	
13	25 Oct		Largest	Arctic	
14	25 Oct		Relatively Large	Arctic	
15	1 Nov		Relatively Low	Siberia	
16	3 Nov		Relatively Low	Siberia	

* Date of announcement, not necessarily shot date.

** Mr. McCone on Oct. 26, 1958, announced that seven of these test detonations have been of high yield, meaning, that each had an explosive power equal to millions of tons of TNT.

1961

1.	1 Sep	Intermediate	Siberia
2.	4 Sep	Intermediate	Siberia
3.	5 Sep	Intermediate	Siberia
4.	6 Sep	Intermediate	East of Stalingrad
5.	10 Sep	Several MT	Arctic
6.	10 Sep	Intermediate	Arctic
7.	12 Sep	Several MT	Arctic
8.	18 Sep	Low-to-Intermediate	Siberia
9.	13 Sep	Low-to-Intermediate	Arctic
10.	14 Sep	Multimegaton	Arctic
11.	16 Sep	Order of 1 MT	Arctic
12.	17 Sep	Intermediate	Siberia
13.	18 Sep	About 1 MT	Arctic
14.	20 Sep	About 1 MT	Arctic
15.	23 Sep	About 1 MT	Arctic
16.	2 Oct	About 1 MT	Arctic
17.	4 Oct	MT Range	Arctic
18.	6 Oct	MT Range	Arctic

U.K. NUCLEAR DETONATIONS

No.	Date	Time (GCT)	Type	Yield	Location
	1952 HURRICANE				
1	3 Oct		Ship	Kiloton Range	Monte Bello Islands
	1953 TOTEM				
1	14 Oct	2230	Tower	Kiloton Range	Woomera {Test held at Emu Field,
2	26 Oct	2230	Tower	Kiloton Range	Woomera {300 miles NW of Woomera.
	1956 MOSAIC				
1	16 May		Tower	Kiloton Range	Monte Bello Islands
2	19 June		Tower	Kiloton Range	Monte Bello Islands
	BUFFALO				
1	27 Sept		Tower	Kiloton Range	Maralinga
2	4 Oct		Surface	Low Yield	Maralinga
3	11 Oct		Air Drop	Low Yield	Maralinga
4	22 Oct		Tower	Kiloton Range	Maralinga
	1957 GRAPPLE				
1	15 May		Air Drop	Megaton Range	Christmas Island Area
2	31 May		Air Drop	Megaton Range	Christmas Island Area
3	19 June		Air Drop	Megaton Range	Christmas Island Area
	ANTLER				
1	14 Sept	0530	Tower	Low Yield	Maralinga
2	25 Sept	0030	Tower	Kiloton Range	Maralinga
3	9 Oct	0730	Balloon	Kiloton Range	Maralinga
	GRAPPLE				
1	8 Nov		Air Drop	Megaton Range	Christmas Island Area
	1958 GRAPPLE				
1	28 Apr		Air Drop	Megaton Range	Christmas Island Area
2	22 Aug		Balloon	Kiloton Range	Christmas Island Area
3	2 Sept		Air Drop	Megaton Range	Christmas Island Area
4	11 Sept		Air Drop	Megaton Range	Christmas Island Area
5	23 Sept		Balloon	Kiloton Range	Christmas Island Area

REPUBLIC OF FRANCE DETONATIONS

	1960				
1	13 Feb	0700	350 ft. Tower	60-70 KT	Reggan
2	1 Apr	0615	Surface	Small	Reggan
3	27 Dec	0730	Tower	Small	Reggan
	1961				
1	25 Apr	0500	Tower	Small	Reggan

APPENDIX IV

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